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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
09/629,482	07/31/2000	Franz Josef Brocker	50487	4024
26474	7590	02/12/2008	EXAMINER	
NOVAK DRUCE DELUCA + QUIGG LLP			DANG, THUAN D	
1300 EYE STREET NW				
SUITE 1000 WEST TOWER			ART UNIT	PAPER NUMBER
WASHINGTON, DC 20005			1797	
			MAIL DATE	DELIVERY MODE
			02/12/2008	PAPER

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

09/629482

Examiner: DANG, THUAN

GAU: 1797

Inventor: BROCKER , FRANZ, et al

Classification: 585/266.000

Status: 124 - ON APPEAL -- AWAITING DECISION BY THE BOARD OF APPEALS

Title: ISOTHERMAL OPERATION OF HETEROGENEOUSLY CATALYZED THREE PHASE REACTIONS

PROSECUTION tab report (75 items, sorted by Date DESC)

Img	Status	Doc Code	Document Type	Date	Pages	Annotations
	10	RBNE	Reply Brief Noted - BPAI	10/17/2007	2	
	7	APRB	Reply Brief Filed	07/31/2007	6	
	7	N417	Status Letter Mailed to Applicant	07/31/2007	2	
	7	APEA	Examiner's Answer to Appeal Brief	05/31/2007	9	
	7	BIB	Bibliographic Data Sheet	05/31/2007	1	
	7	AP.B	Appeal Brief Filed	02/23/2007	18	
	7	WFEE	Fee Worksheet (PTO-06)	02/23/2007	2	
	7	N417	Status Letter Mailed to Applicant	02/23/2007	2	
	7	AP.PRE.DEC	Pre-Brief Appeal Conference decision	01/23/2007	2	
	7	AP.PRE.REQ	Pre-Brief Conference request	12/11/2006	6	
	7	XT/	Extension of Time	12/11/2006	1	
	7	WFEE	Fee Worksheet (PTO-06)	12/11/2006	2	
	7	N417	Status Letter Mailed to Applicant	12/11/2006	2	
	7	N/AP	Notice of Appeal Filed	12/11/2006	1	Appeal no. 2008-1715.
	7	CTAV	Advisory Action (PTOL-303)	10/27/2006	4	
	7	SRFW	Search information including classification, databases and other search related notes	10/27/2006	1	
	7	FWCLM	Index of Claims	10/27/2006	1	
	7	A.NE	Amendment After Final	10/10/2006	1	
	7	REM	Applicant Arguments/Remarks Made in an Amendment	10/10/2006	7	
	7	N417	Status Letter Mailed to Applicant	10/10/2006	2	

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	7	WFEE	Fee Worksheet (PTO-06)	10/10/2006	1	
	7	CTFR	Final Rejection	07/10/2006	7	
	7	SRFW	Search information including classification, databases and other search related notes	07/10/2006	1	
	7	FWCLM	Index of Claims	07/10/2006	1	
	7	PET.DEC.TC	Petition decision routed to the Technology Center Legal Instrument Examiners to act on the decision or continue prosecution.	04/20/2006	1	
	7	PET.OP	Petition for review by the Office of Petitions.	03/16/2006	2	Petition Granted 4/19/06
	7	PETDEC	Petition Decision	03/08/2006	2	
	7	PET.OP	Petition for review by the Office of Petitions.	01/19/2006	2	supplement to 12/19/06 petition
	7	PET.OP	Petition for review by the Office of Petitions.	12/19/2005	2	Petition Dismissed 3/7/06
	7	IMIS	Miscellaneous Internal Document	12/19/2005	3	
	7	ABN	Abandonment	12/08/2005	2	
	7	EXIN	Examiner Interview Summary Record (PTOL - 413)	12/08/2005	2	
	7	A...	Amendment - After Non-Final Rejection	08/12/2004	1	
	7	REM	Applicant Arguments/Remarks Made in an Amendment	08/12/2004	5	
	7	CLM	Claims	08/12/2004	4	
	7	WFEE	Fee Worksheet (PTO-06)	08/12/2004	1	
	7	CTNF	Non-Final Rejection	05/18/2004	6	
	7	FWCLM	Index of Claims	05/18/2004	1	

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Img	Status	Doc Code	Document Type	Date	Pages	Annotations
	7	SRFW	Search information including classification, databases and other search related notes	05/18/2004	1	
	7	SRNT	Examiner's search strategy and results	05/13/2004	1	
	7	RCEX	Request for Continued Examination (RCE)	05/03/2004	2	
	7	REM	Applicant Arguments/Remarks Made in an Amendment	05/03/2004	2	
	7	CLM	Claims	05/03/2004	3	
	7	WFEE	Fee Worksheet (PTO-06)	05/03/2004	1	
	7	N/AP	Notice of Appeal Filed	03/01/2004	1	
	7	CTAV	Advisory Action (PTOL-303)	02/09/2004	3	
	7	ANE.I	Amendment After Final or under 37CFR 1.312, initiated by the examiner.	02/09/2004	1	
	7	A.NE	Amendment After Final	01/14/2004	1	
	7	CLM	Claims	01/14/2004	1	
	7	REM	Applicant Arguments/Remarks Made in an Amendment	01/14/2004	5	
	7	CTFR	Final Rejection	10/29/2003	7	
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	7	REM	Applicant Arguments/Remarks Made in an Amendment	08/22/2003	3	
	7	CLM	Claims	08/22/2003	3	
	7	SPEC	Specification	08/22/2003	11	
	7	CLM	Claims	08/22/2003	3	
	7	ABST	Abstract	08/22/2003	1	

09/629482

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Img	Status	Doc Code	Document Type	Date	Pages	Annotations
	7	IDS	Information Disclosure Statement (IDS) Filed	06/16/2003	2	
	7	CTNF	Non-Final Rejection	05/27/2003	6	
	7	SRNT	Examiner's search strategy and results	05/21/2003	2	
	7	C.AD	Change of Address	04/07/2003	1	
	7	FRPR	Certified Copy of Foreign Priority Application	10/10/2000	21	
	7	CFILE	Request for Corrected Filing Receipt	10/10/2000	4	
	7	TRNA	Transmittal of New Application	07/31/2000	2	
	7	DRW	Drawings-only black and white line drawings	07/31/2000	4	
	7	SPEC	Specification	07/31/2000	11	
	7	CLM	Claims	07/31/2000	3	
	7	ABST	Abstract	07/31/2000	1	
	7	OATH	Oath or Declaration filed	07/31/2000	4	
	7	BIB	Bibliographic Data Sheet	07/31/2000	1	
	7	WFEE	Fee Worksheet (PTO-06)	07/31/2000	1	
	7	WFEE	Fee Worksheet (PTO-06)	07/31/2000	1	
	7	IIFW	Issue Information including classification, examiner, name, claim, renumbering, etc.	07/31/2000	1	
	7	SRFW	Search information including classification, databases and other search related notes	07/31/2000	1	
	7	FWCLM	Index of Claims	07/31/2000	1	

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NOVAK DRUCE DELUCA + QUIGG LLP
1300 EYE STREET NW
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WASHINGTON, DC 20005

Appeal No: 2008-1715
Application: 09/629,482
Appellant: Franz Josef Brocker et al.

Board of Patent Appeals and Interferences Docketing Notice

Application 09/629,482 was received from the Technology Center at the Board on January 25, 2008 and has been assigned Appeal No: 2008-1715.

A review of the file indicates that the following documents have been filed by appellant:

Appeal Brief filed on: February 23, 2007
Reply Brief filed on: July 31, 2007
Request for Hearing filed on: NONE

In all future communications regarding this appeal, please include both the application number and the appeal number.

The mailing address for the Board is:

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The facsimile number of the Board is 571-273-0052. Because of the heightened security in the Washington D.C. area, facsimile communications are recommended. Telephone inquiries can be made by calling 571-272-9797 and should be directed to a Program and Resource Administrator.

By order of the Board of Patent Appeals and Interferences



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BIB DATA SHEET

CONFIRMATION NO. 4024

SERIAL NUMBER	FILING or 371(c) DATE RULE	CLASS	GROUP ART UNIT	ATTORNEY DOCKET NO.
09/629,482		585	1797	50487

APPLICANTS

Franz Josef Brocker, Ludwigshafen, GERMANY;
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 Gerd Kaibel, Lampertheim, GERMANY;
 Gerd Rohrbacher, Ludwigshafen, GERMANY;
 Ekkehard Schwab, Neustadt, GERMANY;
 Manfred Stroezel, Ilvesheim, GERMANY;

**** CONTINUING DATA *********** FOREIGN APPLICATIONS *******

GERMANY 19936276.9 08/02/1999

**** IF REQUIRED, FOREIGN FILING LICENSE GRANTED ****

09/22/2000

Foreign Priority claimed	<input type="checkbox"/> Yes <input type="checkbox"/> No	<input type="checkbox"/> Met after Allowance	STATE OR COUNTRY	SHEETS DRAWINGS	TOTAL CLAIMS	INDEPENDENT CLAIMS
35 USC 119(a-d) conditions met	<input type="checkbox"/> Yes <input type="checkbox"/> No	<input type="checkbox"/> Initials	GERMANY	4	16	2

ADDRESS

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 UNITED STATES

TITLE

Isothermal operation of heterogeneously catalyzed three phase reactions

FILING FEE RECEIVED 690	FEES: Authority has been given in Paper No. _____ to charge/credit DEPOSIT ACCOUNT No. _____ for following:	<input type="checkbox"/> All Fees
		<input type="checkbox"/> 1.16 Fees (Filing)
		<input type="checkbox"/> 1.17 Fees (Processing Ext. of time)
		<input type="checkbox"/> 1.18 Fees (Issue)
		<input type="checkbox"/> Other _____
		<input type="checkbox"/> Credit

Application Number Information

Application Number: 09/629482

Assignments

Filing or 371(c) Date: 07/31/2000 eDan

Effective Date: 07/31/2000

Application Received: 07/31/2000

Patent Number:

Issue Date: 00/00/0000

Date of Abandonment: 00/00/0000

Attorney Docket Number: 50487

Status: 124 /ON APPEAL -- AWAITING DECISION BY THE BOARD
OF APPEALS

Confirmation Number: 4024

Examiner Number: 73604 / DANG, THUAN

Group Art Unit: 1797 IFW Madras

Class/Subclass: 585/266.000

Lost Case: NO

Appeal Number: 20081715

Unmatched Petition: NO

L&R Code: Secrecy Code:1

Third Level Review: NO

Secrecy Order: NO

Status Date: 02/11/2008

Oral Hearing: NO

Title of Invention: ISOTHERMAL OPERATION OF HETEROGENEOUSLY CATALYZED
THREE PHASE REACTIONS

Bar Code	PALM Location	Location Date	Charge to Loc	Charge to Name	Employee Name	Location
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Appln
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✓

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
09/629,482	07/31/2000	Franz Josef Brocker	50487	4024

26474 7590 10/17/2007
NOVAK DRUCE DELUCA & QUIGG, LLP
1300 EYE STREET NW
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WASHINGTON, DC 20005

EXAMINER

DANG, THUAN D

ART UNIT	PAPER NUMBER
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1797

MAIL DATE	DELIVERY MODE
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10/17/2007

PAPER

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APPLICATION NO./ CONTROL NO.	FILING DATE	FIRST NAMED INVENTOR / PATENT IN REEXAMINATION	ATTORNEY DOCKET NO.
09629482	7/31/00	BROCKER ET AL.	50487

EXAMINER

Thuan D.. Dang

ART UNIT	PAPER
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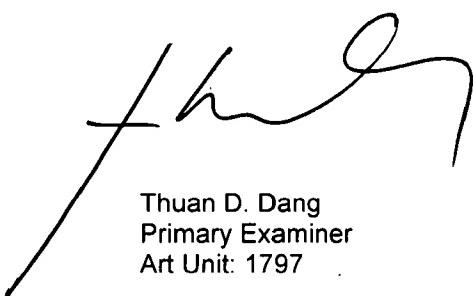
1797 20071009

DATE MAILED:

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Commissioner for Patents

The Reply Brief filed on 7/31/2007 has been entered and sent to the Board of Patent Appeals and Conferences for consideration.



Thuan D. Dang
Primary Examiner
Art Unit: 1797

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:	BRÖCKER ET AL	DOCKET NO.:	50487
SERIAL No.:	09/629,482	CONFIRMATION No.:	4024
FILING DATE:	07/31/2000	EXAMINER:	DANG, THUAN D.
CUSTOMER No.:	26474	ART UNIT:	1764
FOR: ISOTHERMAL OPERATION OF HETEROGENEOUSLY CATALYZED THREE PHASE REACTIONS			

Honorable Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

Reply Brief under 37 C.F.R. § 41.41

Sir:

This is a Reply Brief to the Examiner's Answer mailed May 31, 2007. Please charge any shortage in fees due in connection with the filing of this paper, including Extension of Time fees, to Deposit Account 14.1437. Please credit any excess fees to such account.

Respectfully submitted,
Novak Druce & Quigg, LLP



Michael P. Byrne
Registration No.: 54,015

1300 Eye St. N.W.
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REMARKS**Rejections under 35 U.S.C. §112, second paragraph:**

In the Examiner's Answer mailed May 31, 2007, the examiner states that "[i]t is unclear the expression [sic] 'without ... fluid' is applied to the passing step or the flow in the reactor."

In response, appellants would like to re-emphasize the arguments presented in the Appeal Brief filed February 23, 2007, to the effect that the phrase "passing the generated reaction fluid, without substantial change in the degree of the dispersion of said reaction fluid, through a reactor"¹ has a clear and precise meaning that is readily discernible to one of ordinary skill in the art, especially when read in light of the present specification. Moreover, appellants have clearly asserted that the claim requires there to be substantially no change in the degree of dispersion both in the feed line and through the reactor. Accordingly, Appellants assert that instant claims 11 – 16, are definite and respectfully request reversal of the instant rejection under 35 U.S.C. §112.

Rejections under 35 U.S.C. §103:

Claims 11 – 16 stand rejected under 35 U.S.C. §103(a) over Arganbright et al. (USPN 4,950,834), however, the Examiner has not set forth a *prima facie* case of obviousness with regard to the invention of claim 11 as a whole, because claim 11 requires at least the following:

I. A process for carrying out a reaction under isothermal conditions

Arganbright et al. do not teach a process for carrying out a reaction under isothermal conditions. To the contrary, Arganbright et al. teach that

the temperature along the column will be as in any distillation column, the highest temperature will be in the bottom and the temperature along the column will be the

¹ Claim 11.

boiling point of the composition at that point in the column under the particular conditions of pressure.²

Since, Arganbright et al. do not teach a process for carrying out a reaction under isothermal conditions, and the examiner has provided no apparent reason to modify the reference in this regard, the present rejection is in error and should be reversed.

II. Generating a Reaction Fluid and Passing the Generated Reaction Fluid through a Reactor

Claim 11 requires the step of “generating a reaction fluid by dispersing the gas phase containing the at least one gaseous reactant in the liquid phase, containing the at least one liquid reactant and producing a reaction thereby.” The claim as a whole makes clear that this generation step is not performed inside the reactor, because the generated reaction fluid is subsequently passed through a reactor. As argued in the Appeal Brief filed February 23, 2007, Arganbright et al. not only fails to teach or suggest this claim limitation. The examiner has failed to point to any teaching, suggestion or motivation for a person of ordinary skill in the art to modify the reference to arrive at a process which includes the step of generating a reaction fluid and subsequently passing the generated reaction fluid through a reactor.

III. Passing the Generated Reaction Fluid through a Reactor without substantial change in the degree of dispersion of the reaction fluid

Claim 11 requires the step of “passing the generated reaction fluid, without substantial change in the degree of the dispersion of said reaction fluid, through a reactor.”³ The Arganbright et al. reference provides no apparent reason to avoid substantial change in the degree of dispersion of a reaction fluid as it passes through a reactor. The examiner has not attempted to point to such an apparent reason, but instead merely states that the claimed reactor “is indistinguishable from the one used in the

² Column 8, indicated lines 34 – 39 of US 4,950,834.

³ Claim 11.

Arganbright process.”⁴

On the one hand, the examiner argues that propylene “must be dispersed” into any down-flowing benzene stream, and that therefore some sort of pseudo-reaction fluid is generated in the Arganbright et al. reactor. On the other hand, the examiner argues that this pseudo-reaction fluid passes through the reactor without substantial change in the degree of the dispersion. Unless the examiner also takes the position that the pseudo-reaction fluid is generated instantaneously, both arguments cannot be true. Assuming for the sake of argument that propylene is dispersed into a down-flowing benzene stream, a skilled artisan would not expect such dispersion to occur instantaneously, but would expect the propylene to be dispersed gradually, if at all. If the dispersion occurs gradually, it is clear that the degree of dispersion changes through the reactor, and that the Arganbright et al. reference does not teach “passing the generated reaction fluid, without substantial change in the degree of the dispersion of said reaction fluid, through a reactor.”

IV. Reactor Space Equipped with Woven or Knitted Metal Fabrics Coated with Catalyst

Arganbright et al.’s disclosure of enclosing catalytic particles in a container such as a cloth, screen wire or polymeric mesh in no way teaches or suggests the utilization of a woven or knitted metal fabric coated with a catalyst. The examiner’s assertion that “[c]learly sieves must be on the knitted wire (coated),”⁵ is precisely the opposite of what Arganbright et al. disclosed, i.e. that catalytic particles are enclosed in a container such as a cloth, screen wire or polymeric mesh.

In the Examiner’s Answer mailed May 31, 2007, the examiner states that the reference “discloses … how the catalyst is loaded in the reactor[,]”⁶ and then somehow concludes that “it is clear that the Arganbright reactor is equipped with metal meshes coated with catalysts.” To the contrary, it should be clear that the molecular sieves, “three-dimensional alumina-silicates of the zeolite mineral group,”⁷ of Arganbright et al.,

⁴ Page 6, lines 20 – 21 of the Examiner’s Answer.

⁵ Page 3, lines 1 – 3 of the Advisory Action dated October 27, 2006 (emphasis added).

⁶ Page 7, lines 2 – 4 of the Examiner’s Answer.

⁷ Column 3, indicated lines 65 – 66 of US 4,950,834.

in no way obviate a coating of a woven or knitted metal fabric.

V. Transferring Heat

Claim 11 requires the step of “transferring the heat through the reactor to a cooling fluid medium on the reaction wall surface facing away the reactor space.”⁸ As discussed in the Appeal Brief, filed February 23, 2007, the examiner’s contention that an air conditioned room would be sufficient to control reactor temperature is wholly contrary to the teachings of Arganbright et al., who teach controlling temperature by modifying pressure. Moreover, the Examiner’s contention that an air conditioned room would be sufficient to control reactor temperature is tenuous at best, wholly speculative and completely unreasonable absent objective evidence illustrating the same.

VI. Separating the Reaction Fluid into Gas and Liquid phases

Claim 11 requires the step of “separating the reaction fluid into gas phase and liquid-phase.” As discussed in the Appeal Brief, filed February 23, 2007, in the Arganbright et al. process “the initial reaction product is removed from the reaction zone as quickly as it is formed,”⁹ flowing to the bottom of the distillation column and exiting via line 8, and

[t]he feed of propylene is adjusted such that there is a molar excess of benzene in the reactor, such that the overhead 5 is primarily benzene, the propylene having been almost totally reacted. In addition to benzene and some propylene other lights go off overhead. The overhead is passed to condenser 13 which is operated to condense substantially all of the benzene¹⁰

Thus, contrary to the examiner’s statement that the claimed step “must be recognized when on[e] having ordinary skill in the art see [sic] figure 1 in which unreacted materials as a gas go out [sic] the reactor via stream 5 and cumene as a liquid flow down stream

⁸ Claim 11.

⁹ Column 3, lines 24 – 25 of Arganbright et al. (US 4,950,834).

¹⁰ Column 7, lines 37 – 44 of Arganbright et al. (US 4,950,834) (emphasis in original).

8[,]”¹¹ Arganbright et al. never disclose “separating [a] reaction fluid into gas phase and liquid-phase.”¹²

In Conclusion:

The examiner has failed to establish a *prima facie* case of obviousness. For at least these reasons, the rejection of claim 11 is in error and should be reversed. “If an independent claim is nonobvious under 35 U.S.C. 103, then any claim depending therefrom is nonobvious.”¹³ Thus, claims 12 – 16 are also nonobvious.

¹¹ Page 7, lines 16 – 18 of the Examiner’s Answer.

¹² Claim 11.

¹³ MPEP §2143.03, citing *In re Fine*, 837 F.2d 1071, 5 USPQ2d 1596 (Fed. Cir. 1988).



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**BEFORE THE BOARD OF PATENT APPEALS
AND INTERFERENCES**

Application Number: 09/629,482

Filing Date: July 31, 2000

Appellant(s): BROCKER ET AL.

MAILED
MAY 31 2007
GROUP 1700

Michael Byrne
For Appellant

EXAMINER'S ANSWER

This is in response to the appeal brief filed 2/23/2007 appealing from the Office action mailed 7/10/2006.

(1) Real Party in Interest

A statement identifying by name the real party in interest is contained in the brief.

(2) Related Appeals and Interferences

The examiner is not aware of any related appeals, interferences, or judicial proceedings which will directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

(3) Status of Claims

The statement of the status of claims contained in the brief is correct.

(4) Status of Amendments After Final

The appellant's statement of the status of amendments after final rejection contained in the brief is correct.

(5) Summary of Claimed Subject Matter

The summary of claimed subject matter contained in the brief is correct.

(6) Grounds of Rejection to be Reviewed on Appeal

The appellant's statement of the grounds of rejection to be reviewed on appeal is correct.

(7) Claims Appendix

The copy of the appealed claims contained in the Appendix to the brief is correct.

(8) Evidence Relied Upon

4,950,834

ARGANBRIGHT et al.

8-1990

(9) Grounds of Rejection

The following ground(s) of rejection are applicable to the appealed claims:

Claim Rejections - 35 USC § 112

The following is a quotation of the second paragraph of 35 U.S.C. 112:

The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.

Claims 11-16 are rejected under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.

Regarding claim 11, the “passing step” is indefinite due to the recitation of the limitation “without substantial change in the degree of the dispersion of said reaction fluid” since it is unclear the change in the degree of dispersion is applied thru the **entire** the catalyst bed or just in the connection path between the “generating zone” the reactor.

Claim Rejections - 35 USC § 103

The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

(a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

The factual inquiries set forth in *Graham v. John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103(a) are summarized as follows:

1. Determining the scope and contents of the prior art.
2. Ascertaining the differences between the prior art and the claims at issue.
3. Resolving the level of ordinary skill in the pertinent art.
4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

This application currently names joint inventors. In considering patentability of the claims under 35 U.S.C. 103(a), the examiner presumes that the subject matter of the various claims was commonly owned at the time any inventions covered therein were made absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and invention dates of each claim that was not commonly owned at the time a later invention was made in order for the examiner to consider the applicability of 35 U.S.C. 103(c) and potential 35 U.S.C. 102(e), (f) or (g) prior art under 35 U.S.C. 103(a).

Claims 11-16 are rejected under 35 U.S.C. 103(a) as obvious over Arganbright et al (4,950,834).

Arganbright discloses a process of reacting between propylene and benzene in the presence of a catalyst substantially the same as the applicant's claimed catalyst in an isothermal reactor having a wall contacted with the surrounding air (the abstract; the drawings; column 1, lines 8-66; col. 3, lines 1, lines 1-60; col. 5, line 14 thru col. 6, line 35).

As disclosed on column 3, line 28, benzene in the reactor is boiling (a liquid form).

Arganbright discloses that propylene is the most volatile component in the reaction (col. 8, lines 3-4). Arganbright also discloses that the reaction includes both vapor and liquid (col. 8, lines 52-55). Therefore, propylene must inherently be in the form of gas before the reaction.

On column 7, lines 24-34, Arganbright discloses that the benzene and others flow down to the bottom of the Omega sieve section to the Y sieve section. In figure 1 and 2, the examiner has recognized that gas propylene stream 1 must be dispersed into this flowing down benzene stream before the mixture of benzene and propylene reacted further in the Y sieve.

Arganbright does not disclose that (1) the mixture of benzene and propylene does not substantially change in the degree of dispersion thru the reactor (Y sieve section) and (2) using a cooling fluid medium for delivering the heat away from the reactor.

However, it is expected that the dispersion would not be change in the Y sieve section of the Arganbright process since the catalyst bed of the Arganbright reactor is made by the same material as the claimed reactor (col. 5, line 14 thru col. 6, line 55).

It is expected that the heat of the reaction will be transfer from the wall of the reactor via the surrounding air.

However, in the case that the reaction room is too hot, it would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Arganbright process by employing air conditioners to cool down the room to arrive at the applicants' claimed process to avoid the reaction room is too hot.

A recycle of benzene can be found in figures.

Temperature and pressure can be found on column 8, lines 42-46.

Arganbright appears to be silent as to the superficial liquid/gas velocity. However, these parameters depend on the size of the reactor and selected conversion.

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Arganbright process by selecting appropriate velocities of gas/liquid to operate the process since it is expected that using any superficial liquid/gas velocity would yield similar results.

(10) Response to Argument

Regarding the argument of the rejection under 35 USC 112, 2nd paragraph, the examiner maintains the same position as maintained in the Advisory Action mailed on 10/27/2006. It is unclear the expression “without . . . fluid” is applied to the passing step or the flow in the reactor. Although applicants maintain that in the specification, the change of the dispersion is applied to both the feed line and through the reactor, the claim is clear on its face. The limitation of the claim must be correctly interpreted so that any difference between the prior art and the claimed process can be recognized.

The argument that there is not intentional step of generating a reaction fluid by dispersing a gas phase in a liquid phase is not persuasive since as shown in figure 1 and in the abstract, propylene gas stream 1 must be dispersed into the downstream benzene (stream 2) before the mixture benzene and propylene reacted further in Y sieve zone. Appellants also argue that as described in the specification, the dispersing step of gas phase in the liquid phase can be performed by a dispersing elements such as a liquid jet gas compressor, jet pumps for conveying and compressing gasses. However, applicants do not claim using these dispersing elements.

The argument that it is not simply the material of construction , but overall design of the reaction and catalyst which influence whether a substantial change in the degree of the dispersion of the reaction fluid occurs as it passes through the reactor is not persuasive since appellants do not claim which structure in the design make an unchanged dispersion possible. Instead, appellants claim “through a reactor whose reactor space is equipped with woven or knitted metal fabrics coated with catalyst”. This reactor equipped with these elements is indistinguishable from the one used in the Arganbright process as discussed in the above rejection and the later respond.

The argument that no way the reference teaches or suggests the utilization of a woven or knitted metal fabric coated with a catalyst is not persuasive since Arganbright discloses everywhere from column 5, lines 14 through column 7, line 5 about how the catalyst is loaded in the reactor, namely column 6, lines 8-12; column 6, line 62 through column 7, line 1. Appellants also argue that catalyst particles are not enclosed in containers in the present invention, while the prior art discloses the catalyst are enclosed in a container. However, it does not matter if the catalyst in a container or not, it is clear that the Arganbright reactor is equipped with metal meshes coated with catalysts.

The argument that as described in the specification, in the present process, the heat transfer is provided by heat exchangers comprising a plate type heat exchanger or spiral heat exchanger is not persuasive since appellants do not claim using exchangers. Further, as understood from the law of nature, heat must be transferred from a hot region to a cooler region. Therefore, there must be a transfer of heat between the reactor and the surrounds and this heat transfer must be via the reactor wall.

The argument that claim 11 requires a step of ‘separating the reaction fluid into gas phase and liquid phase’. This step must be recognized when one having ordinary skill in the art see figure 1 in which unreacted materials as a gas go out the reactor via stream 5 and cumene as a liquid flow down stream 8.

(11) Related Proceeding(s) Appendix

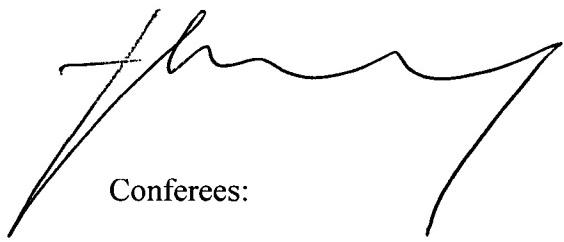
No decision rendered by a court or the Board is identified by the examiner in the Related Appeals and Interferences section of this examiner’s answer.

Art Unit: 1764

For the above reasons, it is believed that the rejections should be sustained.

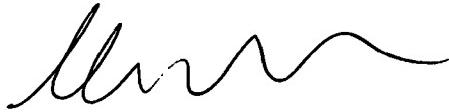
Respectfully submitted,

Thuan D Dang



Conferees:

Glenn Calderola



Glenn Calderola
Supervisory Patent Examiner
Technology Center 1700



Kathryn Gorgos

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:	BRÖCKER ET AL	DOCKET NO.:	50487
SERIAL No.:	09/629,482	CONFIRMATION No.:	4024
FILING DATE:	07/31/2000	EXAMINER:	DANG, THUAN D.
CUSTOMER No.:	26474	ART UNIT:	1764
FOR: ISOTHERMAL OPERATION OF HETEROGENEOUSLY CATALYZED THREE PHASE REACTIONS			

Honorable Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

Appeal brief under 37 C.F.R. § 41.37

Sir:

This is an appeal from the Examiner's final rejection of Claims 11 - 16, dated July 10, 2006. Claims 1 - 16 are currently pending.

The fee set forth in 37 C.F.R. § 41.20(b)(2) is paid by credit card. Form PTO-2038 is enclosed. Please charge any shortage in fees due in connection with the filing of this paper, including Extension of Time fees, to Deposit Account 14.1437. Please credit any excess fees to such account.

Respectfully submitted,
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REAL PARTY IN INTEREST:

The real party in interest is BASF Aktiengesellschaft, of Ludwigshafen, Germany.

RELATED APPEALS AND INTERFERENCES:

To the best of the undersigned's knowledge, there are no related interferences or judicial proceedings within the meaning of 37 C.F.R. §1.192(c).

STATUS OF CLAIMS:

- Claims 1 - 16 are pending in the application
- Claims 11 - 16 stand rejected, and are being appealed.
- Claims 1 - 10 stand withdrawn from consideration.

STATUS OF AMENDMENT:

No amendment was filed subsequent to the final rejection dated July 10, 2006.

SUMMARY OF CLAIMED SUBJECT MATTER

The claimed invention relates to a process for the isothermal operation of heterogeneously catalyzed reactions involving at least three phases in the form of a gaseous phase, a liquid phase and a solid phase.¹

The independent claim involved in the appeal is claim 11. All other claims are dependent on claim 11. Summary of the subject matter of the dependent claims is omitted as unnecessary.

¹ Page 1, lines 9 – 12 of the Specification.

Claim 11 is directed to a process for carrying out a catalyzed three-phase reaction under isothermal conditions.² The three phases involved in this reaction are a gas phase, containing at least one gaseous reactant, a liquid phase containing at least one liquid reactant and a solid phase which is a catalyst.³ The process of claim 11 comprises four steps. In the first step, a reaction fluid is generated by dispersing the gas phase in the liquid phase.⁴ Subsequently, the generated reaction fluid is passed through a reactor.⁵ The reactor space is equipped with woven or knitted metal fabrics coated with catalyst.⁶ Care is taken to ensure that no substantial change in the degree of the dispersion of the reaction fluid occurs as it passes through the reactor.⁷ Heat is transferred through the reactor to a cooling fluid medium on the reaction wall surface facing away the reactor space.⁸ Finally the reaction fluid is separated into gas phase and liquid-phase.⁹

GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

- Whether the examiner erred in rejecting Claims 11-16 under 35 U.S.C. §112, second paragraph as allegedly being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention, and
- Whether the examiner erred in rejecting Claims 11 – 16 under 35 U.S.C. §103(a) as allegedly being obvious over Arganbright et al. (USPN 4,950,834).

ARGUMENT

Rejections under 35 U.S.C. §112, second paragraph:

Claims 11-16 stand rejected as allegedly being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention. The examiner states that the phrase, “passing the generated reaction fluid, without substantial change in the degree of the dispersion of said reaction fluid, through a

² Page 1, lines 9 – 12 of the Specification.

³ Page 1, lines 14 – 17 of the Specification.

⁴ Page 3, lines 27 – 28 of the Specification.

⁵ Page 7, lines 9 – 10 of the Specification.

⁶ Page 3, lines 34 – 35 and Page 4, line 28 – Page 5, line 34 of the Specification.

⁷ Page 4, lines 8 – 26 of the Specification.

⁸ Page 6, lines 16 – Page 7, line 2 of the Specification.

⁹ Page 7, lines 14 – 17 of the Specification.

reactor^{”¹⁰ is indefinite on the basis that:}

it is unclear the change [sic] in the degree of dispersion is applied thru [sic] the **entire** the [sic] catalyst bed or just in the connection path between the ‘generating zone’ the [sic] reactor.¹¹

In other words, the examiner argues that the phrase “passing the … fluid, without substantial change in the degree of dispersion … through a reactor”¹² would be misinterpreted by one possessing ordinary skill in the pertinent art. More specifically, the examiner argues that a skilled artisan would not be reasonably apprised that appellants have claimed a process in which the degree of dispersion is not substantially changed throughout the entire reactor. The examiner argues that a skilled artisan would interpret the phrase “passing the … fluid, without substantial change in the degree of dispersion … through a reactor”¹³ as signifying that the degree of dispersion of the fluid could change when it passes through the reactor and that appellants have actually claimed a process in which the degree of dispersion only remains substantially unchanged through “the connection path between the generating zone [and] the reactor.”¹⁴

This rejection is in error. “The requirement to ‘distinctly’ claim means that the claim must have a meaning discernible to one of ordinary skill in the art when construed according to correct principles.... Only when a claim remains insolubly ambiguous without a discernible meaning after all reasonable attempts at construction must a court declare it indefinite.”¹⁵ Moreover, “[i]n rejecting a claim under the second paragraph of 35 U.S.C. 112, it is incumbent on the examiner to establish that one of ordinary skill in the pertinent art, when reading the claims in light of the supporting specification, would not have been able to ascertain with reasonable degree of precision and particularity the particular area set out and circumscribed by the claims.”¹⁶

It is respectfully submitted that the phrase “passing the generated reaction fluid, without substantial change in the degree of the dispersion of said reaction fluid, through a

¹⁰ Claim 11.

¹¹ Page 2, lines 10 – 12 of the final Office action of July 10, 2006 (emphasis in original).

¹² Claim 11.

¹³ Claim 11.

¹⁴ Page 2, lines 10 – 12 of the final Office action of July 10, 2006 (emphasis in original).

¹⁵ *Metabolite Labs., Inc. v. Lab. Corp. of Am. Holdings*, 370 F.3d 1354, 1366, 71 USPQ2d 1081, 1089 (Fed. Cir. 2004).

¹⁶ *Ex parte Wu*, 10 USPQ2d 2031 at 2033 (BPAI 1989).

reactor”¹⁷ has a clear and precise meaning that is readily discernible to one of ordinary skill in the art, especially when read in light of the present specification, which states:

The inventors have determined that improved mass transfer can only be obtained if the reaction fluid is a dispersion formed from the gas phase (as disperse phase) and the liquid (as dispersion medium) and the process and apparatus are designed in such a way that the dispersion, as it passes through the reactor, remains stable, ie. substantially no increase in bubble size occurs.¹⁸

In this regard, the specification also points to particular features of the reactor, stating:

The reactor of the invention is designed for maintaining a high but uniform shearing stress on the reaction fluid. On the other hand, it will withstand a high cross-sectional flow velocity without attrition of the catalyst. On the other, the reaction fluid is exposed to a uniformly high shearing stress in the metal fabric. This provides for uniform mixing of the reaction fluid and hence for a constant degree of dispersion of the reaction fluid as it passes through the reactor.”¹⁹

Accordingly, Appellants assert that instant claims 11 – 16, are definite and respectfully request reversal of the instant rejection under 35 U.S.C. §112.

Rejections under 35 U.S.C. §103:

Claims 11 – 16 stand rejected under 35 U.S.C. §103(a) as allegedly being obvious over Arganbright et al. (USPN 4,950,834). Appellants respectfully submit that the Examiner has failed to establish a *prima facie* case of obviousness under 35 U.S.C. §103. When applying 35 U.S.C. § 103, the following tenets of patent law must be adhered to: (A) The claimed invention must be considered as a whole; (B) The references must be considered as a whole and must suggest the desirability and thus the obviousness of making the combination; (C) The references must be viewed without the benefit of impermissible hindsight vision afforded by the claimed invention; and (D) Reasonable expectation of success is the standard with which obviousness is determined.²⁰

¹⁷ Claim 11.

¹⁸ Page 4, lines 8 – 15 of the Specification.

¹⁹ Page 4, lines 17 – 26 of the Specification.

²⁰ *Hodosh v. Block Drug Co., Inc.*, 786 F.2d 1136, 1143 n.5, 229 USPQ 182, 187 n.5 (Fed. Cir. 1986).

“To establish a *prima facie* case of obviousness, three basic criteria must be met. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings.”²¹ Indeed, “to support the conclusion that the claimed invention is directed to obvious subject matter, either the references must expressly or impliedly suggest the claimed invention or the examiner must present a convincing line of reasoning as to why the artisan would have found the claimed invention to have been obvious in light of the teachings of the references.”²² The examiner may not, because of doubt that the invention is patentable, resort to speculation, unfounded assumption or hindsight reconstruction to supply deficiencies in the factual basis for the rejection.²³ “Second, there must be a reasonable expectation of success.”²⁴ The teaching or suggestion to make the claimed combination and the reasonable expectation of success must both be found in the prior art, and not based on applicant’s disclosure.²⁵ That it would have been obvious to try is not sufficient.²⁶ “Finally, the prior art reference (or references when combined) must teach or suggest all the claim limitations.”²⁷

In view of the above principles, the Examiner has not set forth a *prima facie* case of obviousness with regard to the invention of claim 11 as a whole, because claim 11 requires at least the following:

- I. Generating a Reaction Fluid
- II. Passing the Generated Reaction Fluid through a Reactor (without substantial change in the degree of dispersion of the reaction fluid)
- III. Reactor Space Equipped with Woven or Knitted Metal Fabrics Coated with Catalyst
- IV. Transferring Heat
- V. Separating the Reaction Fluid into Gas and Liquid phases

²¹ MPEP §2143.

²² *Ex parte Clapp*, 227 USPQ 972, 973 (Bd. Pat. App. & Inter. 1985).

²³ See *In re Warner*, 379 F.2d 1011, 1017, 154 USPQ 173, 177 (CCPA 1967), cert. denied, Appeal No. 2002-1187 389 U.S. 1057 (1968).

²⁴ MPEP §2143. See also: *Boehringer Ingelheim Vetmedica, Inc. v. Schering-Plough Corp.*, 320 F.3d 1339, 1354 (Fed. Cir. 2003).

²⁵ *In re Vaeck*, 947 F.2d 488, 20 USPQ2d 1438 (Fed. Cir. 1991).

²⁶ *Hybritech, Inc. v. Monoclonal Antibodies, Inc.*, 802 F.2d 1367, 1380 (Fed. Cir. 1986).

²⁷ MPEP §2143.

These requirements will be addressed one at a time, because a *prima facie* case of obviousness cannot be established if the cited reference fails to teach or suggest any one of the claim limitations, however, it is important to bear in mind that the claimed invention must be considered as a whole. Indeed, this invention is greater than the sum of its parts.

I. Generating a Reaction Fluid

Claim 11 requires the step of “generating a reaction fluid by dispersing the gas phase containing the at least one gaseous reactant in the liquid phase, containing the at least one liquid reactant and producing a reaction thereby.” As described in the specification, dispersing of the gas phase in the liquid phase can be performed by a dispersing element, such as a liquid jet gas compressor, e.g., jet pumps for conveying and compressing gasses. In complete contrast, Arganbright et al. describe a dual bed system wherein a feed through the reactor is accomplished by separate feed streams.²⁸ Moreover, Arganbright et al. actively seek to avoid what they consider to be “the detriment of having all of the components of the reaction system continually in contact with the catalyst,”²⁹ emphasizing that “because [their] reaction is occurring concurrently with distillation, the initial reaction product is removed from the reaction zone as quickly as it is formed.”³⁰ Thus, it is clear that the reference not only fails to teach or suggest all of the claim limitations, but strenuously teaches away from generating a reaction fluid which could continually contact the catalyst. The examiner has failed to point to any teaching, suggestion or motivation for a person of ordinary skill in the art to modify the reference to arrive at a process which includes the step of generating a reaction fluid. Despite these clear differences, the Examiner asserts that:

“Arganbright discloses that the benzene and others flow to the bottom of the Omega sieve section to the Y sieve section.”³¹

²⁸ Col. 3, lines 1-5 of Arganbright et al. (US 4,950,834).

²⁹ Col. 3, lines 61 – 63 of Arganbright et al. (US 4,950,834).

³⁰ Col. 3, lines 23 – 25 of Arganbright et al. (US 4,950,834).

³¹ Page 3 at lines 22 – 23 of the final Office action of July 10, 2006.

The examiner then speculates that:

“gas propylene stream 1 must be dispersed into this flowing down benzene stream before the mixture of benzene and propylene reacted further in the Y sieve.”³²

However, the Examiner has considered the cited passage in a vacuum and has failed to consider the teachings of the reference as a whole. Indeed, the complete passage recited by Arganbright et al. do not describe an active process for generating a reaction fluid. Arganbright et al specifically state, “[s]ince complete separation of the reaction products and benzene does not occur in Omega bed 12, cumene, small amounts of propylene, dimerized propylene...and benzene flow down the column into the Y-bed where any propylene and dimerized propylene may react with the benzene to produce additional cumene.”³³ This passage does not describe a step for generating a reaction fluid. Products of a reaction and left-over reactants are merely allowed to flow to the bottom of the column. No intentional step of generating a reaction fluid by dispersing a gas phase in a liquid phase is described. Moreover, contrary to the examiner’s speculation, Arganbright et al. do not teach that the small amounts of propylene “must be dispersed” into any down-flowing benzene stream. In fact, Arganbright et al., teach away from this possibility by stating that additional reaction, if any, occurs in the Y-bed, i.e. by stating that “[products of a reaction and left-over reactants] ... flow down the column into the Y-bed where any propylene and dimerized propylene may react with the benzene to produce additional cumene.”³⁴ This passage clearly does not provide a teaching suggestion or motivation for a skilled artisan to intentionally generate a reaction fluid by dispersing a gas phase into a liquid phase.

In sum, the reference fails to teach or suggest all of the claim limitations, but instead teaches away from generating a reaction fluid. Where the prior art “teaches away” from the claimed invention rather than motivating a person of ordinary skill in the art to do what the patentee has done, the claimed invention is nonobvious.³⁵ The

³² Page 4 at lines 1 – 2 of the final Office action of July 10, 2006.

³³ Col. 7, lines 24-34 of Arganbright et al. (US 4,950,834).

³⁴ Col. 7, lines 24-34 of Arganbright et al. (US 4,950,834).

³⁵ *In re Hedges*, 783 F.2d 1038, 1041 (Fed. Cir. 1986); *W.L. Gore & Assocs. v. Garlock, Inc.*, 721 F.2d

examiner has failed to point to any teaching, suggestion or motivation for a person of ordinary skill in the art to modify the reference to arrive at a process which includes the step of generating a reaction fluid. A *prima facie* case of obviousness has not been established. For at least these reasons, the rejection of claim 11 is in error and should be reversed. “If an independent claim is nonobvious under 35 U.S.C. 103, then any claim depending therefrom is nonobvious.”³⁶ Thus, claims 12 – 16 are also nonobvious.

II. Passing the Generated Reaction Fluid through a Reactor without substantial change in the degree of dispersion of the reaction fluid

Claim 11 also requires the step of “passing the generated reaction fluid, without substantial change in the degree of the dispersion of said reaction fluid, through a reactor.”³⁷ First, since Arganbright et al. teach away from generating a reaction fluid (as discussed above), it is unreasonable to suggest that Arganbright et al. teach or suggest the passing of a generated reaction fluid through a reactor. The examiner has ignored this impossibility, but has, at least, admitted that:

“Arganbright does not disclose that … the mixture of benzene and propylene does not substantially change in the [sic] degree of dispersion thru [sic] the reactor (Y sieve section)”³⁸

The examiner further speculates that:

“the dispersion would not be change [sic] in the Y sieve section of the Arganbright process since the catalyst bed of the Arganbright reactor is made by the same material as the claimed reactor.”³⁹

It is noted that for the above-mentioned reasons, the examiner’s use of the phrase “mixture of benzene and propylene” and the term “dispersion” is entirely inappropriate. It has already been noted that the examiner may not resort to speculation, unfounded

³⁶ 1540, 1552-53 (Fed. Cir. 1983).

³⁷ MPEP §2143.03, citing *In re Fine*, 837 F.2d 1071, 5 USPQ2d 1596 (Fed. Cir. 1988).

³⁸ Claim 11.

³⁹ Page 4 at lines 3 – 5 of the final Office action of July 10, 2006.

³⁹ Page 4 at lines 6 – 8 of the final Office action of July 10, 2006 (emphasis added).

assumption or hindsight reconstruction to supply deficiencies in the factual basis for the rejection. Aside from being purely speculative, the examiner's assertion is wholly unreasonable because the present application makes clear that it is not simply the material of construction, but the overall design of the reactor and catalyst which influence whether a substantial change in the degree of the dispersion of the reaction fluid occurs as it passes through the reactor. The present invention discloses a reactor designed for maintaining a high but uniform shearing stress on the reaction fluid. This design provides for uniform mixing of the reaction fluid and a constant degree of dispersion of the reaction fluid as it passes through the reactor.⁴⁰

The Arganbright et al. reference provides no teaching, suggestion or motivation to create a reaction fluid; no teaching, suggestion or motivation to avoid substantial change in the degree of dispersion of a reaction fluid as it passes through a reactor; and, quite naturally, no teaching suggestion or motivation as to how to arrive at a process wherein the overall design of the reactor and catalyst influence whether a substantial change in the degree of the dispersion of a reaction fluid occurs as it passes through a reactor. The Arganbright et al. reference, therefore, fails to teach or suggest all of the claim limitations, and the examiner has failed to establish a *prima facie* case of obviousness. For at least these reasons, the rejection of claim 11 is in error and should be reversed. "If an independent claim is nonobvious under 35 U.S.C. 103, then any claim depending therefrom is nonobvious."⁴¹ Thus, claims 12 – 16 are also nonobvious.

III. Reactor Space Equipped with Woven or Knitted Metal Fabrics Coated with Catalyst

Claim 11 requires that the reaction fluid be passed "through a reactor whose reactor space is equipped with woven or knitted metal fabrics coated with catalyst."⁴² The examiner's initial rejection did not point to a teaching, suggestion or motivation for a skilled artisan to have arrived at this feature of the present invention. In response to remarks presented by the appellants, however, the examiner has stated that:

⁴⁰ Page 4, indicated lines 17 – 26 of the Specification.

⁴¹ MPEP §2143.03, citing *In re Fine*, 837 F.2d 1071, 5 USPQ2d 1596 (Fed. Cir. 1988).

⁴² Claim 11.

Arganbright discloses that the molecular sieve can be enclosed in the screen wire which is knitted. Clearly sieves must be on the knitted wire (coated).⁴³

The examiner has failed to examine the Arganbright et al. reference as a whole, and has instead resorted to cherry-picking distinct aspects from the reference to construct the rejection. The Federal Circuit in *Hodosh v. Block Drug Co., Inc.*, 786 F.2d 1136, 1143, n.5 (Fed. Cir. 1986) stated that "references must be considered as a whole and must suggest the desirability and thus the obviousness of making the combination." In this regard, the Examiner examined only parts of the cited art references picking and choosing only disclosures allegedly favorable for establishing *prima facie* obviousness.

Arganbright et al. discuss the use of mole sieve catalysts. The examiner cites column 5, lines 20 – 22, which state: "the particulate mole sieves may be employed by enclosing them in a porous container such as a cloth, screen wire or polymeric mesh." Even on the basis of this hand-selected portion of Arganbright et al., the examiner has failed to establish a *prima facie* case of obviousness; a reference disclosing the enclosure of catalytic particles in a container such as a cloth, screen wire or polymeric mesh in no way teaches or suggests the utilization of a woven or knitted metal fabric coated with a catalyst. The examiner's assertion that "[c]learly sieves must be on the knitted wire (coated),"⁴⁴ is precisely the opposite of what Arganbright et al. disclosed, i.e. that catalytic particles are enclosed in a container. Catalytic particles are not enclosed in containers in the present invention. The present invention effectively makes woven or knitted metal fabrics catalytically active, by coating them with catalyst. When properly considered as a whole, the Arganbright et al. reference teaches away from such a configuration. The examiner cherry-picked Column 5, lines 20 – 22, but the next two lines make clear that "[t]he material used to make the container must be inert to the reactants and conditions in the reaction system."⁴⁵ Thus, the Arganbright et al. reference fails to teach or suggest all of the claim limitations, because it fails to teach woven or knitted metal fabrics coated with catalyst. Moreover, Arganbright et al., teach away from making such a modification to their system. The examiner has failed to establish a *prima*

⁴³ Page 3, lines 1 – 3 of the Advisory Action dated October 27, 2006.

⁴⁴ Page 3, lines 1 – 3 of the Advisory Action dated October 27, 2006 (emphasis added).

⁴⁵ Column 5, lines 23 – 24 of Arganbright et al. (US 4,950,834).

facie case of obviousness. For at least these reasons, the rejection of claim 11 is in error and should be reversed. “If an independent claim is nonobvious under 35 U.S.C. 103, then any claim depending therefrom is nonobvious.”⁴⁶ Thus, claims 12 – 16 are also nonobvious.

IV. Transferring Heat

Claim 11 requires the step of “transferring the heat through the reactor to a cooling fluid medium on the reaction wall surface facing away the reactor space.”⁴⁷ With regard to this required step, the examiner has admitted that:

“Arganbright does not disclose … using a cooling fluid medium for delivering heat away from the reactor.”⁴⁸

Yet, the examiner speculates that:

“it is expected that the heat of the reaction will be transfer [sic] from the wall of the reactor via the surrounding air.”⁴⁹

The examiner further speculates that:

“it would have been obvious to one having ordinary [sic] skill in the art at the time the invention was made to have modified the Arganbright process by employing air conditioners to cool down the room to arrive at the applicants’ claimed process to avoid the reaction room is too hot [sic].”⁵⁰

Again, the examiner may not resort to speculation, unfounded assumption or hindsight reconstruction to supply deficiencies in the factual basis for the rejection. Moreover, as will be pointed out, the examiner’s assertion is wholly contrary to the teachings of Arganbright et al., who specifically teach controlling temperature by modifying pressure.

⁴⁶ MPEP §2143.03, citing *In re Fine*, 837 F.2d 1071, 5 USPQ2d 1596 (Fed. Cir. 1988).

⁴⁷ Claim 11.

⁴⁸ Page 4 at lines 3 – 5 of the final Office action of July 10, 2006.

⁴⁹ Page 4 at lines 9 – 10 of the final Office action of July 10, 2006.

⁵⁰ Page 4 at lines 11 – 14 of the final Office action of July 10, 2006 (emphasis added).

In the present invention, heat transfer is provided by a heat exchanger, for example, as described in the specification as comprising a plate type heat exchanger or a spiral type heat exchanger, which, as illustrated in FIG. 4, utilizes a counter-current flow, albeit co-current arrangements are also disclosed. On the other hand, Arganbright does not disclose cooling systems, e.g., countercurrent type heat exchangers, but rather, discloses, “[t]he temperature of the reactor is determined by boiling point of the liquid mixture...To change the temperature the pressure is changed. Temperature in the reaction zone is thus controlled by pressure; by increasing the pressure, the temperature in the system is increased, and vice versa.”

Accordingly, not only has the Examiner failed to consider the teachings of Arganbright as a whole (Arganbright teaches pressure not an air conditioned room), but the Examiner's proposed modification is wholly contrary to the teachings of Arganbright. Indeed, Arganbright specifically teaches controlling temperature by virtue of modifying pressure. Furthermore, there is simply no reasonable expectation that the combination/modification propounded by the Examiner would be successful; that is, the Examiner's contention that an air conditioned room would be sufficient to control reactor temperature, or be sufficient to cool the instant claimed reactor is tenuous at best, wholly speculative and completely unreasonable absent objective evidence illustrating the same.

Since the Arganbright et al. reference fails to teach or suggest all of the claim limitations the examiner has failed to establish a *prima facie* case of obviousness. Additionally, Arganbright et al. teach away from making the examiner's proposed modification of cooling via an air conditioned room by pointing towards controlling the temperature via pressure. Where the prior art “teaches away” from the claimed invention rather than motivating a person of ordinary skill in the art to do what the patentee has done, the claimed invention is nonobvious.⁵¹ Finally, a skilled artisan would not have reasonably expected the modifications proposed by the examiner to be successful. Thus, the examiner has failed to establish a *prima facie* case of obviousness. For at least these reasons, the rejection of claim 11 is in error and should be reversed. “If an independent

⁵¹ *In re Hedges*, 783 F.2d 1038, 1041 (Fed. Cir. 1986); *W.L. Gore & Assocs. v. Garlock, Inc.*, 721 F.2d 1540, 1552-53 (Fed. Cir. 1983).

claim is nonobvious under 35 U.S.C. 103, then any claim depending therefrom is nonobvious.”⁵² Thus, claims 12 – 16 are also nonobvious.

V. Separating the Reaction Fluid into Gas and Liquid phases

Claim 11 requires the step of “separating the reaction fluid into gas phase and liquid-phase.” First, since Arganbright et al. teach away from generating a reaction fluid (as discussed above), it is unreasonable to suggest that Arganbright et al. teach or suggest separating a reaction fluid into gas and liquid phases. Moreover, the Arganbright et al. reference is directed to catalytic distillation. Arganbright et al. specifically note that:

The success of catalytic distillation lies in an understanding of the principles associated with distillation. First, because the reaction is occurring concurrently with distillation, the initial reaction product is removed from the reaction zone as quickly as it is formed Third, the reaction has an increased driving force because the reaction products have been removed and cannot contribute to reverse reaction (Le Chatelier’s Principle).⁵³

The examiner has failed to establish a *prima facie* case of obviousness, because the examiner has failed to point to a teaching, suggestion or motivation to modify the disclosure of Arganbright et al. to arrive at a process which includes a step for “separating the reaction fluid into gas phase and liquid-phase.”⁵⁴

As already mentioned, the Arganbright et al. process teaches away from generating a reaction fluid in the first place. Moreover, in the Arganbright et al. process “the initial reaction product is removed from the reaction zone as quickly as it is formed,”⁵⁵ flowing to the bottom of the distillation column and exiting via line 8, and

[t]he feed of propylene is adjusted such that there is a molar excess of benzene in the reactor, such that the overhead 5 is primarily benzene, the propylene having been almost totally reacted. In addition to benzene and some propylene

⁵² MPEP §2143.03, citing *In re Fine*, 837 F.2d 1071, 5 USPQ2d 1596 (Fed. Cir. 1988).

⁵³ Column 3, lines 19 – 35 of Arganbright et al. (US 4,950,834).

⁵⁴ Claim 11.

⁵⁵ Column 3, lines 24 – 25 of Arganbright et al. (US 4,950,834).

other lights go off overhead. The overhead is passed to condenser 13 which is operated to condense substantially all of the benzene⁵⁶

Thus, Arganbright et al. never disclose “separating [a] reaction fluid into gas phase and liquid-phase[,]”⁵⁷ and, the examiner has failed to establish a *prima facie* case of obviousness. For at least these reasons, the rejection of claim 11 is in error and should be reversed. “If an independent claim is nonobvious under 35 U.S.C. 103, then any claim depending therefrom is nonobvious.”⁵⁸ Thus, claims 12 – 16 are also nonobvious.

⁵⁶ Column 7, lines 37 – 44 of Arganbright et al. (US 4,950,834) (emphasis in original).

⁵⁷ Claim 11.

⁵⁸ MPEP §2143.03, citing *In re Fine*, 837 F.2d 1071, 5 USPQ2d 1596 (Fed. Cir. 1988).

CLAIMS APPENDIX.

1. (withdrawn) Apparatus for carrying out reactions involving a gaseous phase, a and a solid phase being a catalyst, which comprises the steps of, comprising
 - a dispersing element (6) for dispersing a gas phase in a liquid phase to generate a reaction fluid,
 - at least one reactor (1) which possesses an inlet (31, 41), an outlet (43) and a reactor space bounded by heat-removing walls which are spaced apart substantially uniformly along the main flow axis of the reaction fluid, and which is fitted with catalyst-coated metal fabric (20,32), and
 - a feed line (7) which routes the reaction fluid from the dispersing element (6) to the reactor inlet (31,41) and is sufficiently short that the degree of dispersion of the reaction fluid does not substantially change in the course of the passage through the feed line.
2. (withdrawn) Apparatus as claimed in claim 1, wherein the metal fabric (20,32) is knitted metal fabric.
3. (withdrawn) Apparatus as claimed in claim 1, wherein the metal fabric (20,32) is knitted metal fabric.
4. (withdrawn) Apparatus as claimed in claim1, wherein the dispersing element (6) is a liquid jet gas compressor.
5. (withdrawn) Apparatus as claimed in claim 1, wherein the reactor (1) is constructed as a heat exchanger.
6. (withdrawn) Apparatus as claimed in claim 5, wherein the reactor (1) is constructed as a plate type heat exchanger.
7. (withdrawn) Apparatus as claimed in claim 5, wherein the reactor (1) is constructed as a spiral type heat exchanger.
8. (withdrawn) Apparatus as claimed in claim 5, wherein the walls in the reactor are spaced from 1 to 3 mm apart.
9. (withdrawn) Apparatus as claimed in claim 5, wherein the walls in the reactor are spaced from 2 to 20 mm apart.

10. (withdrawn) Apparatus as claimed in claim 5, wherein the walls in the reactor are spaced from 4 to 10 mm apart.
11. (previously presented) A process for carrying out a reaction under isothermal conditions involving a gas phase, containing at least one gaseous reactant, a liquid phase containing at least one liquid reactant and a solid phase which is a catalyst, which comprises the steps of
 - generating a reaction fluid by dispersing the gas phase containing the at least one gaseous reactant in the liquid phase, containing the at least one liquid reactant and producing a reaction thereby,
 - passing the generated reaction fluid, without substantial change in the degree of the dispersion of said reaction fluid, through a reactor whose reactor space is equipped with woven or knitted metal fabrics coated with catalyst
 - transferring the heat through the reactor to a cooling fluid medium on the reaction wall surface facing away the reactor space, and
 - separating the reaction fluid into gas phase and liquid-phase.
12. (original) A process as claimed in claim 11, operated with separate partial recycling of gas phase and/or liquid phase.
13. (original) A process as claimed in claim 11, wherein the superficial liquid velocity in the reactor is from 100 to 66 m³.m⁻²h).
14. (original) A process as claimed in claim 11, wherein the superficial gas velocity from 0.5 to 15 cm/s.
15. (original) A process as claimed in claim 11, wherein the reaction fluid in the reactor is under a pressure of from 0.1 to 200 bar.
16. (original) A process as claimed in claim 11, wherein the reaction fluid in the reactor has a temperature of from 25 to 250°C.

EVIDENCE APPENDIX

None.

RELATED PROCEEDINGS APPENDIX

None.



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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
09/629,482	07/31/2000	Franz Josef Brocker	50487	4024
26474	7590	07/10/2006	EXAMINER	
NOVAK DRUCE DELUCA & QUIGG, LLP			DANG, THUAND	
1300 EYE STREET NW			ART UNIT	PAPER NUMBER
SUITE 400 EAST TOWER				
WASHINGTON, DC 20005			1764	

DATE MAILED: 07/10/2006

Please find below and/or attached an Office communication concerning this application or proceeding.

Office Action Summary	Application No.	Applicant(s)	
	09/629,482	BROCKER ET AL.	
	Examiner	Art Unit	
	Thuan D. Dang	1764	

– The MAILING DATE of this communication appears on the cover sheet with the correspondence address –

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

- 1) Responsive to communication(s) filed on 12 August 2004.
- 2a) This action is **FINAL**. 2b) This action is non-final.
- 3) Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

- 4) Claim(s) 1-16 is/are pending in the application.
- 4a) Of the above claim(s) 1-10 is/are withdrawn from consideration.
- 5) Claim(s) _____ is/are allowed.
- 6) Claim(s) 11-16 is/are rejected.
- 7) Claim(s) _____ is/are objected to.
- 8) Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

- 9) The specification is objected to by the Examiner.
- 10) The drawing(s) filed on _____ is/are: a) accepted or b) objected to by the Examiner.
Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).
Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
- 11) The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

Priority under 35 U.S.C. § 119

- 12) Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
 - a) All b) Some * c) None of:
 1. Certified copies of the priority documents have been received.
 2. Certified copies of the priority documents have been received in Application No. _____.
 3. Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

* See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

- | | |
|---|---|
| 1) <input type="checkbox"/> Notice of References Cited (PTO-892) | 4) <input type="checkbox"/> Interview Summary (PTO-413) |
| 2) <input type="checkbox"/> Notice of Draftsperson's Patent Drawing Review (PTO-948) | Paper No(s)/Mail Date. _____ |
| 3) <input type="checkbox"/> Information Disclosure Statement(s) (PTO-1449 or PTO/SB/08)
Paper No(s)/Mail Date _____. | 5) <input type="checkbox"/> Notice of Informal Patent Application (PTO-152) |
| | 6) <input type="checkbox"/> Other: _____. |

DETAILED ACTION

Claim Rejections - 35 USC § 112

The following is a quotation of the second paragraph of 35 U.S.C. 112:

The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.

Claims 11-16 are rejected under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.

Regarding claim 11, the “passing step” is indefinite due to the recitation of the limitation “without substantial change in the degree of the dispersion of said reaction fluid” since it is unclear the change in the degree of dispersion is applied thru the entire the catalyst bed or just in the connection path between the “generating zone” the reactor.

Claim Rejections - 35 USC § 103

The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

(a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

The factual inquiries set forth in *Graham v. John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103(a) are summarized as follows:

1. Determining the scope and contents of the prior art.

Art Unit: 1764

2. Ascertaining the differences between the prior art and the claims at issue.
3. Resolving the level of ordinary skill in the pertinent art.
4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

This application currently names joint inventors. In considering patentability of the claims under 35 U.S.C. 103(a), the examiner presumes that the subject matter of the various claims was commonly owned at the time any inventions covered therein were made absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and invention dates of each claim that was not commonly owned at the time a later invention was made in order for the examiner to consider the applicability of 35 U.S.C. 103(c) and potential 35 U.S.C. 102(e), (f) or (g) prior art under 35 U.S.C. 103(a).

Claims 11-16 are rejected under 35 U.S.C. 103(a) as obvious over Arganbright et al (4,950,834).

Arganbright discloses a process of reacting between propylene and benzene in the presence of a catalyst substantially the same as the applicant's claimed catalyst in an isothermal reactor having a wall contacted with the surrounding air (the abstract; the drawings; column 1, lines 8-66; col. 3, lines 1, lines 1-60; col. 5, line 14 thru col. 6, line 35).

As disclosed on column 3, line 28, benzene in the reactor is boiling (a liquid form).

Arganbright discloses that propylene is the most volatile component in the reaction (col. 8, lines 3-4. Arganbright also discloses that the reaction includes both vapor and liquid (col. 8, lines 52-55). Therefore, propylene must inherently be in the form of gas before the reaction.

On column 7, lines 24-34, Arganbright discloses that the benzene and others flow down to the bottom of the Omega sieve section to the Y sieve section. In figure 1 and 2, the examiner

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has recognized that gas propylene stream 1 must be dispersed into this flowing down benzene stream before the mixture of benzene and propylene reacted further in the Y sieve.

Arganbright does not disclose that (1) the mixture of benzene and propylene does not substantially change in the degree of dispersion thru the reactor (Y sieve section) and (2) using a cooling fluid medium for delivering the heat away from the reactor.

However, it is expected that the dispersion would not be change in the Y sieve section of the Arganbright process since the catalyst bed of the Arganbright reactor is made by the same material as the claimed reactor (col. 5, line 14 thru col. 6, line 55).

It is expected that the heat of the reaction will be transfer from the wall of the reactor via the surrounding air.

However, in the case that the reaction room is too hot, it would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Arganbright process by employing air conditioners to cool down the room to arrive at the applicants' claimed process to avoid the reaction room is too hot.

A recycle of benzene can be found in figures.

Temperature and pressure can be found on column 8, lines 42-46.

Arganbright appears to be silent as to the superficial liquid/gas velocity. However, these parameters depend on the size of the reactor and selected conversion.

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Arganbright process by selecting appropriate velocities of gas/liquid to operate the process since it is expected that using any superficial liquid/gas velocity would yield similar results.

Response to Arguments

Applicant's arguments filed 8/12/2004 have been fully considered but they are not persuasive.

The argument that as clearly defined in claim 1, lines 21-27, the feed stream line (7) which routes the reaction fluid from dispersing element (6) to the reactor inlet (31,41) has to be sufficiently short so that of the degree of the dispersion of the reaction fluid does not substantially change in the course of the passage through the feed line, now in claim 11 is not persuasive since applicants does not claim using any dispersing element, does not claim using a short reactor inlet and how short the inlet should be.

The argument that the present invention proposed the use of a catalyst-coated metal fabric. See on column 5, lines 20-22 and 55-57, Arganbright discloses that the molecular sieve can be enclosed in screen wire which is knitted. Clearly, sieves must be on the knitted wire (coated). Therefore, there is no difference between the catalyst bed of the Arganbright reactor and the catalyst used in the claimed process.

THIS ACTION IS MADE FINAL. Applicant is reminded of the extension of time policy as set forth in 37 CFR 1.136(a).

A shortened statutory period for reply to this final action is set to expire THREE MONTHS from the mailing date of this action. In the event a first reply is filed within TWO MONTHS of the mailing date of this final action and the advisory action is not mailed until after the end of the THREE-MONTH shortened statutory period, then the shortened statutory period will expire on the date the advisory action is mailed, and any extension fee pursuant to 37

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CFR 1.136(a) will be calculated from the mailing date of the advisory action. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the mailing date of this final action.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Thuan D. Dang whose telephone number is 571-272-1445. The examiner can normally be reached on Mon-Thu.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Glenn Calderola can be reached on 571-272-1444. The fax phone number for the organization where this application or proceeding is assigned is 703-872-9306.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

Thuan D. Dang
Primary Examiner
Art Unit 1764

09629482.20060630
June 30, 2006

A handwritten signature in black ink, appearing to read "Thuan D. Dang".



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WASHINGTON, DC 20005

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MAR 08 2006

In re Application of
Franz Josef Brocker et al.
Application No. 09/629,482
Filed: July 31, 2000
Attorney Docket No.: 50487

OFFICE OF PETITIONS

ON PETITION

This is a decision on the petition filed December 19, 2005 and supplemented on January 19, 2005, to withdraw the holding of abandonment for the above-identified application. The petition is treated under 37 CFR 1.181.

The petition under 37 CFR 1.181 is **DISMISSED**.

Any request for reconsideration of this decision must be submitted within **TWO (2) MONTHS** from the mail date of this decision. Extensions of time under 37 CFR 1.136(a) are permitted. The reconsideration request should include a cover letter entitled "Renewed Petition under 37 CFR 1.181".

The application was held abandoned on August 19, 2004, for failure to file a timely reply to the non-Final Office Action mailed May 18, 2004. A three month period for reply was set. Accordingly, a Notice of Abandonment was mailed on December 8, 2005.

Petitioner asserts that a response was filed August 12, 2004. In support, petitioner has submitted, *inter alia*, a their filing log a copy of the search results from the Patent Application Information Retrieval system both showing an entry for an amendment on August 12, 2005. Additionally, petitioners have submitted a cover page used for submitting the response with a certificate of mail of August 12, 2004.

A search of the application file and the USPTO records does not disclose that the actual amendment is of record. It appears, from the proof submitted though that petitioner attempted to file the response in a timely manner but until such time as a proper response, the amendment is of record, the petition cannot be granted and the abandonment withdrawn.

Please submit the amendment, not just the cover page with a renewed petition for further consideration.

Further correspondence with respect to this matter should be addressed as follows:

By mail: Mail Stop Petition
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

By FAX: (571) 273-8300

Telephone inquiries concerning this matter may be directed to the undersigned
Petitions Attorney at (571) 272-3212.



Patricia Faison-Ball
Senior Petitions Attorney
Office of Petitions

FIG.1

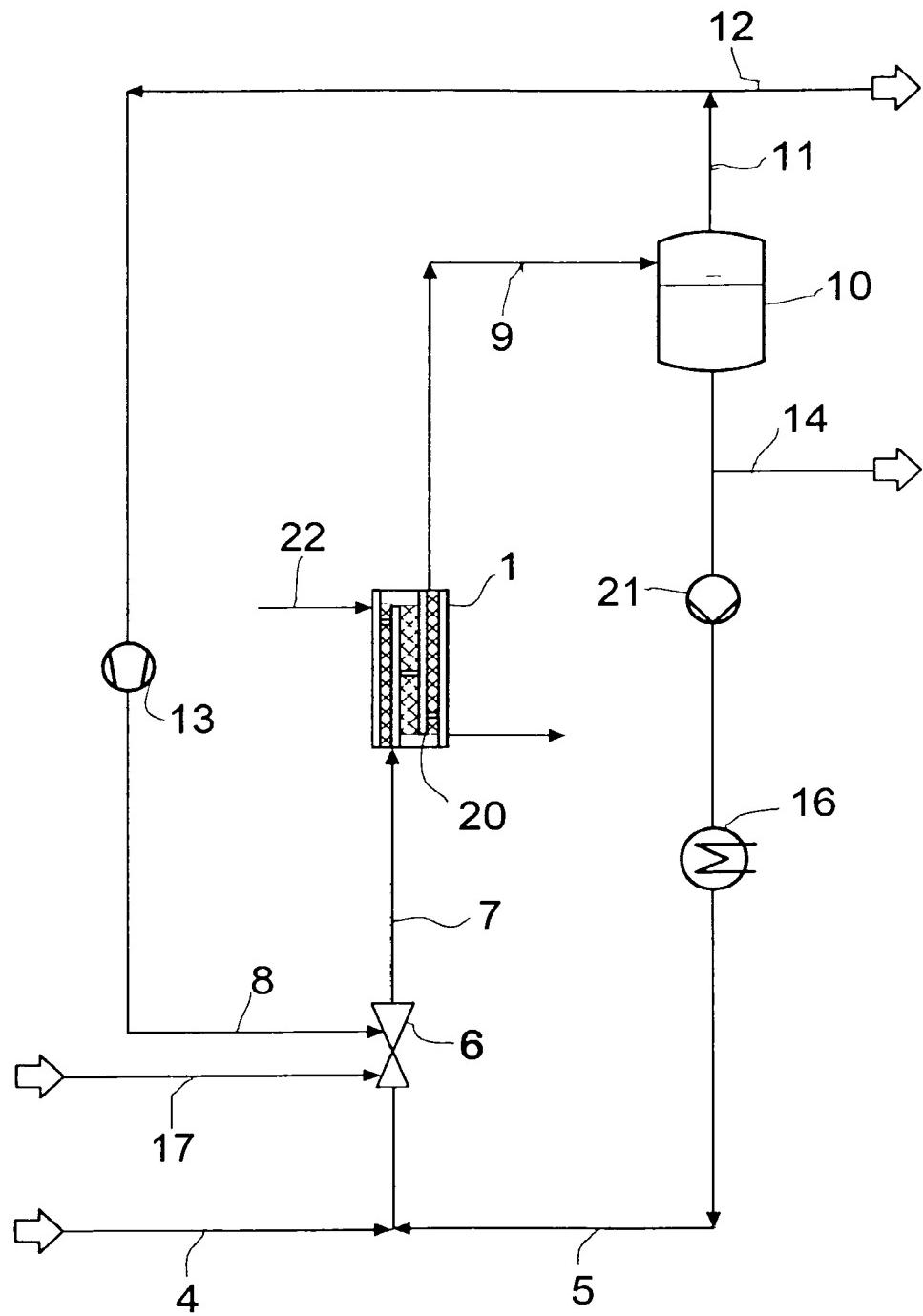


FIG.2

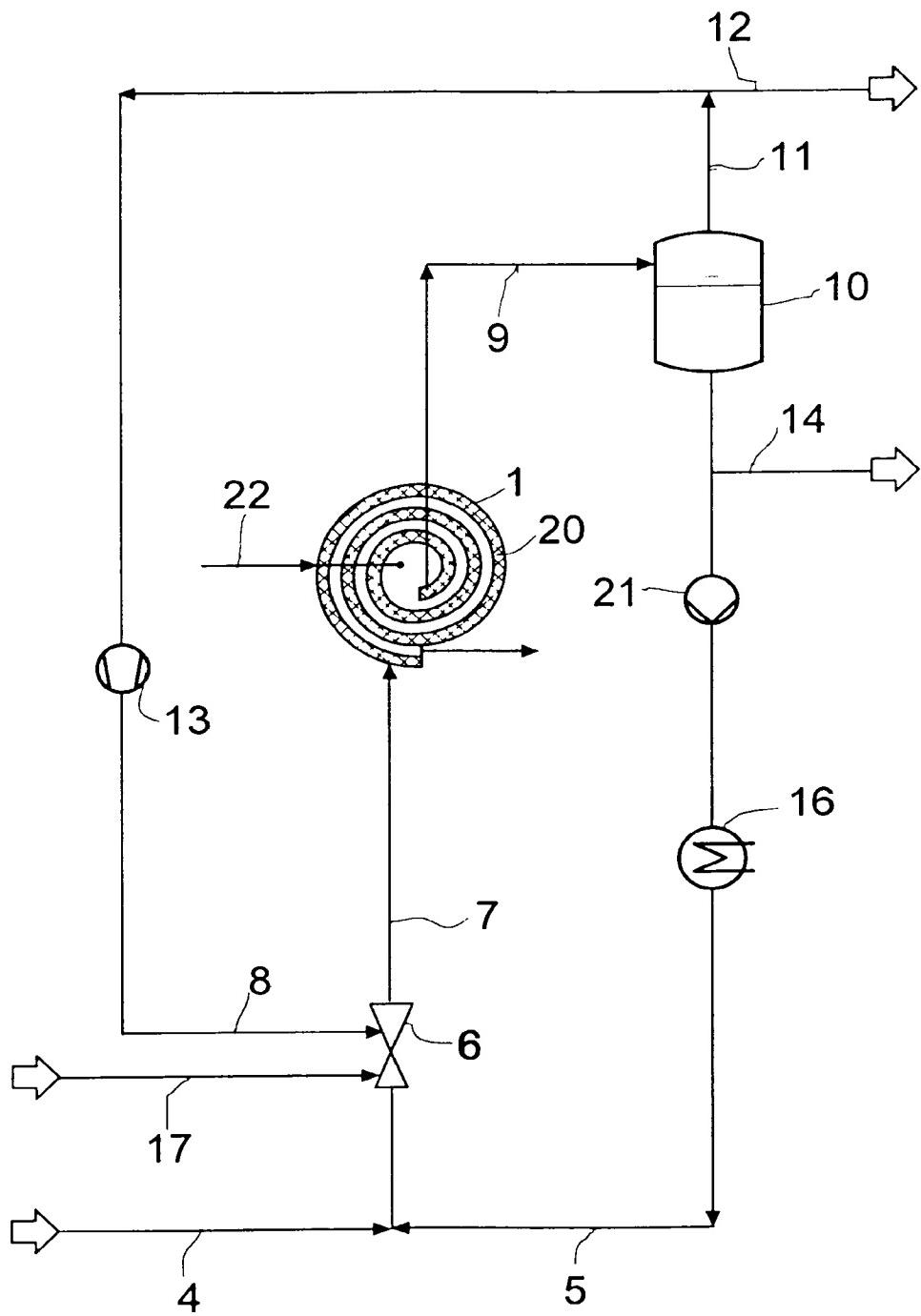


FIG.3

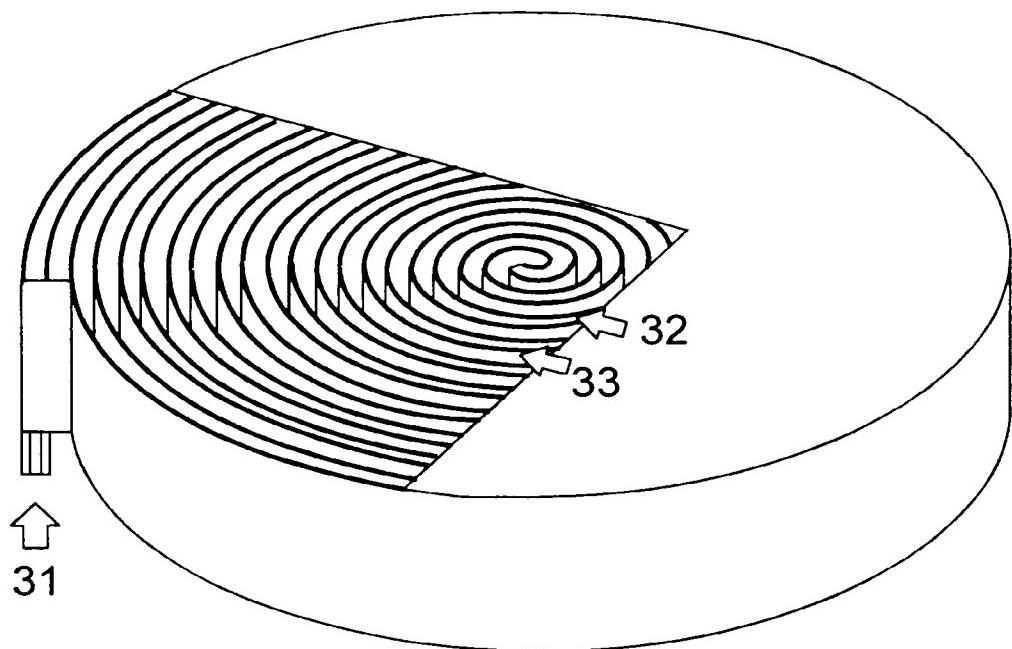
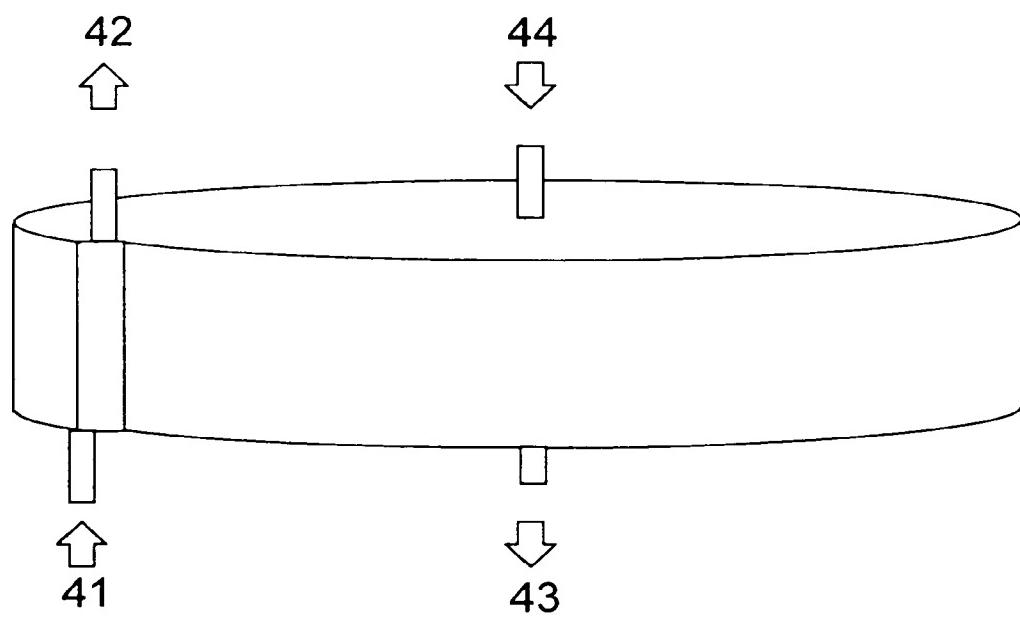


FIG.4



5

Isothermal operation of heterogeneously catalyzed three phase reactions

This invention relates to a process and apparatus for 10 the isothermal operation of heterogeneously catalyzed reactions involving at least three phases in the form of a gaseous phase, a liquid phase and a solid phase.

The invention relates specifically to the operation of 15 such reactions where at least one reactant is liquid and one is gaseous and where the catalyst is a solid material.

The operation of such reactions is associated with 20 appreciable difficulties. The gas-liquid transfer is frequently problematical. Moreover, isothermal conditions are difficult to achieve, isothermal being used in the sense that the heat of reaction is substantially evened out by input or removal of heat, so that temporal or local temperature fluctuations in 25 the reactor are of no consequence.

Established processes are described in G. Eigenberger, Ullmann, 5th edition, vol. 4, p. 199ff. (1992) Wiley-VCH, Weinheim, Berlin, New York.

30

EP-B 0 305 203 (US-A 4 985 230) describes the operation of heterogeneously catalyzed reactions under nonadiabatic conditions. To this end, a reactor with heat-transmitting walls is packed with monolithic catalysts. A monolithic catalyst is a coherent solid having a sufficiently large catalytic surface area that countable amounts of these bodies will suffice for 35

catalyzing the reaction in question to an industrially sensible extent. The monolithic catalysts have channels which are angled relative to the overall flow direction, so that the reaction fluid is routed at an acute angle from one reactor wall to the other, which is said to improve the heat transfer. The shearing stress exerted on the reaction fluid is extremely high (high pressure drop) in reactor wall vicinity and otherwise rather low (poor mass transfer). This leads to unnecessarily large pressure drops in wall vicinity. The reactor is complicated to fabricate, since the pressure drop depends decisively on the geometry between reactor wall and monolithic catalyst.

15 EP-B 0 201 614 (US-A 4 731 229) describes a reactor containing partly corrugated tape-form catalyst bodies whose corrugation is disposed at an inclination to the main flow axis and oppositely directed in adjacent plates, the pitch of the corrugation of the catalyst body being less than the pitch of the adjacent corrugated plates and the surface area of the catalyst body being larger than the surface area of an adjacent corrugated plate. This apparatus is not contemplated for generating gas-liquid dispersions. The complicated corrugation of the plates favors bypass formation, inhibits eddying and thus compromises mass transfer. In addition, the envisioned compact packing element does not provide for effective removal of the heat of reaction.

30 EP-B 0 068 862 (CA-A 1 146 148) discloses a fixed bed reactor for transfer reactions between gas phase and liquid. In this reactor, the fixed bed comprises alternating layers of plane and corrugated sheets coiled together to form a roll, the corrugated sheet comprising an open mesh material with at least an outer surface layer consisting of a high molecular weight

organic polymeric substance which will be inherently hydrophobic with respect to the liquid mentioned, and the plane sheet comprising knitted, woven or felted cloth of a textile wicking material which is 5 hydrophilic with respect to the liquid or the gas-liquid transfer reaction and which will provide an uninterrupted wicking path between the ends of the roll for the liquid mentioned. The disadvantage with this 10 type of reactor is that the textile constituents of the reactor limit the cross-sectional flow velocities. The wicking path, moreover, inhibits rapid liquid transport, thus favors the separation between gas and liquid and inhibits the mass transfer between gas and liquid. Besides, the reactor is intended for adiabatic 15 operation.

It is an object of the present invention to provide apparatus and a process for carrying out reactions involving a liquid phase, a gaseous phase and a solid 20 phase with improved mass transfer between gas phase and liquid and with isothermal processing.

We have found that this object is achieved by apparatus for carrying out reactions involving a gaseous phase, a 25 liquid phase and a solid phase, comprising

- a dispersing element for dispersing a gas phase in a liquid phase to generate a reaction fluid,
- 30 - at least one reactor which possesses an inlet, an outlet and a reactor space bounded by heat-removing walls which are spaced apart substantially uniformly along the main flow axis of the reaction fluid, and which is fitted with catalyst-coated metal fabric, and
- 35

- a feed line which routes the reaction fluid from the dispersing element to the reactor inlet and is sufficiently short that the degree of dispersion of the reaction fluid does not substantially change in the course of the passage through the feed line.

The inventors have determined that improved mass transfer can only be obtained if the reaction fluid is a dispersion formed from the gas phase (as disperse phase) and the liquid (as dispersion medium) and the process and apparatus are designed in such a way that the dispersion, as it passes through the reactor, remains stable, ie. substantially no increase in bubble size occurs.

The reactor of the invention is designed for maintaining a high but uniform shearing stress on the reaction fluid. On the one hand, it will withstand a high cross-sectional flow velocity without attrition of the catalyst. On the other, the reaction fluid is exposed to a uniformly high shearing stress in the metal fabric. This provides for uniform mixing of the reaction fluid and hence for a constant degree of dispersion of the reaction fluid as it passes through the reactor.

The catalyst-coated metal fabric of the invention is a woven or knitted metal fabric. The wire diameter is generally in the range from 0.01 to 5.0 mm, preferably from 0.04 to 1.0 mm. The mesh size may be varied within wide limits.

These wovens or knits may be coated by the process described in EP-B 0 564 830 (CA-A 2 090 930) or EP-A 0 965 384. EP-B 0 564 830 does not expressly describe the coating of metal knits with catalyst, but

they shall be treated in the same way as woven metal fabrics. For the purposes of the present invention, knitted metal fabrics are metal fabrics formed from one continuous metal thread. Woven metal fabrics, in 5 contrast, are fabrics formed from at least two metal threads.

The coating of woven or knitted metal fabrics with catalysts may also be effected by conventional dip 10 processes, for example according to the process described in EP-A 0 056 435.

If the metal forming the woven or knitted metal fabric is itself catalytically active (possibly after a 15 treatment), coating may be dispensed with entirely.

Woven or knitted metal fabrics may be used in the form of tapes. The catalyst-coated woven and knitted metal fabrics may be corrugated by means of a toothed wheel 20 roll. The introduction of corrugated woven or knitted metal fabric in the reactor makes it possible to alter the packing density of the woven or knitted metal fabric. For instance, a plurality of layers of corrugated and smooth woven or knitted metal fabric may 25 be introduced into the reactor space. Similarly, inert metal sheets may be inserted between layers of woven and knitted metal fabric. In any event, the catalyst-coated woven or knitted metal fabrics must be introduced in such a way that the reactor space is very 30 uniformly packed between the heat-conducting walls. Uniform packing suppresses bypass formation and supports the conduction of heat to the heat-removing reactor walls, which in turn enable the reaction to be carried out under isothermal conditions.

35

In a further embodiment, the dispersing element is a liquid jet gas compressor. These conventional

dispersing means are jet pumps for conveying and compressing gases.

In jet pumps, the jet of driving liquid breaks up into individual droplets on exit from the driving nozzle. These droplets become uniformly distributed across the cross section of the mixing nozzle, entrain ambient gas by impact and friction and compress it to a higher pressure. The attainable degree of dispersion is determined by the setting of driving nozzle and diffuser. This in turn depends on the pressure of the driving liquid, the suction pressure, the counterpressure, the flow of driving liquid, the gas suction stream and the mixture stream.

In a further embodiment of the apparatus according to the invention, the reactor is constructed as a heat exchanger. The transmission of heat through the reactor wall is decisively increased when a fluid medium on the reactor wall surface facing away from the reactor space takes up the heat of reaction and carries it away. Such a heat exchanger reactor can be constructed as a plate type heat exchanger or as a spiral type heat exchanger. A plate type heat exchanger reactor according to the invention has an in particular square or rectangular reactor space which is subdivided by additional heat-conducting walls which force the reaction fluid to take a zigzag course through the reactor space. Where the change of direction is greatest no catalyst-coated woven or knitted metal fabric is used in order that an excessively large pressure drop may be avoided. A spiral type heat exchanger reactor according to the invention has an in particular cylindrical reactor space which is packed very uniformly with catalyst-coated woven or knitted metal fabrics. The wall spacing of the heat exchanger reactors of the invention is

preferably from 1 to 30 mm, especially from 2 to 20 mm, in particular from 4 to 10 mm.

The invention further provides a process for carrying 5 out reactions involving a gaseous phase, a liquid phase and a solid phase, which comprises the steps of

- generating a reaction fluid by dispersing a gas phase in a liquid phase,
- passing the generated reaction fluid through a 10 reactor whose reactor space is fitted with woven or knitted metal fabrics coated with catalyst,
- transferring the heat of reaction at the walls which bound the reactor space, and
- separating the reaction fluid into gas phase and 15 liquid phase.

The separating of the reaction fluid may be effected using conventional separators.

The process is preferably carried out with the overall 20 direction of flow of reaction fluid in the reactor being upward.

A further embodiment of the process according to the invention is operated with separate partial recycling 25 of gas phase and/or liquid phase. By separate partial recycling is meant that the reaction product is separated from the gas phase and/or from the liquid phase. The remaining gas and the remaining liquid may be completely or partially redispersed and fed back to 30 the reactor.

In a further embodiment of the process according to the invention, the superficial liquid velocity in the reactor is from 100 to 600 m³/(m²·h), preferably from 35 150 to 300 m³/(m²·h). The superficial liquid velocity is the volume flow of the liquid fraction of the dispersion at the reaction conditions (pressure and

temperature divided by the cross-sectional area of the reactor space perpendicularly to the main flow axis. Since, as a result of woven or knitted metal fabrics being introduced, the reactor space is not available to 5 the reaction fluid in its entirety, the actual microscopic superficial liquid velocity is correspondingly higher.

In a further embodiment of the process according to the 10 invention, the superficial gas velocity is from 0.5 to 15 cm/s, preferably from 2.5 to 10 cm/s. The superficial gas velocity is herein defined similarly to the superficial liquid velocity.

15 In a further embodiment of the process according to the invention, the reaction fluid in the reactor is under a pressure of from 0.1 to 200 bar, preferably from 1 to 100 bar, especially from 1 to 10 bar.

20 In a further embodiment of the process according to the invention, the reaction fluid in the reactor has a temperature of from 25 to 250°C, preferably from 25 to 200°C, in particular from 50 to 150°C.

25 The invention will now be more particularly described with reference to Figures 1 to 4.

Fig. 1 shows an apparatus for a three phase 30 reaction with product recycling, cycle gas operation using a liquid jet gas compressor and a plate type heat exchanger reactor.

Fig. 2 shows an apparatus for a three phase 35 reaction with product recycling, cycle gas operation using a liquid jet gas compressor and a spiral type heat exchanger reactor.

Fig. 1 shows a side view of the interior of a spiral type heat exchanger reactor.

Fig. 4 shows a side view of a spiral type heat
5 exchanger reactor.

Figs. 1 and 2 show an apparatus which, the metal fabric
20 supported catalyst in the reactor 1 having been
activated (for example, by reduction with H₂), is filled
10 with product liquid by using the circulating pump 21 to
pump the liquid from the separator 10 via the optional
preheater 16 and the feed line to the liquid jet gas
compressor 5 to the liquid jet gas compressor 6 and
from there to the heat exchanger reactor 1 and from it
15 via the feed line to the separator 9 back into the
separator 10. Cycle gas is withdrawn from the separator
10 via the feed line 11 and fed by means of the cycle
gas pump 13 via the feed line to the liquid jet gas
compressor 8 to the liquid jet gas compressor 6, where
20 the gas is compressed and simultaneously dispersed in
the liquid to form the reaction mixture. A sufficiently
short feed line to reactor 7 such that the degree of
dispersion of the reaction fluid does not substantially
alter over this distance is used to feed the reaction
25 fluid into the reactor 1. Once the circulation has been
started up with product, the feed line to the liquid
jet gas compressor 4 is used to introduce reactant, and
a constant fill level system on the separator 10 is
used to withdraw a corresponding amount of product from
30 the liquid circulation via the discharge line 14. Fresh
gas to replace the reaction gas consumed is fed into
the gas circulation via the feed line to the liquid jet
gas compressor 17, with the pressure being maintained,
and off-gas is withdrawn from the gas circulation via
35 the off-gas line 12. In the case of exothermic
reactions the heat of reaction is removed from the

reactor via the cooling circulation system 22, while in the case of endothermic reactions it is introduced.

Fig. 3 shows a side view of a spiral type heat exchanger reactor according to the invention. 31 identifies the feed for the reaction fluid into the reactor (reactor inlet). 32 identifies the reactor passage which will receive the catalyst-coated metal fabric, which will take up the entire space in more or less dense packing. 33 identifies the cooling passage, which is to receive the cooling fluid.

Fig. 4 is a side view of a spiral type heat exchanger reactor and identifies the arrangement of the feed and discharge stubs. 41: reaction fluid feed (reactor inlet), 42: cooling fluid discharge, 43: reaction fluid discharge (reactor outlet), 44: cooling fluid feed. Reaction fluid and cooling fluid are here arranged in countercurrent in order that the heat transfer may be maximized. If the amount of heat released at the reactor inlet specifically is critical with regard to, for example, selectivity and catalyst stability, then a cocurrent arrangement is advisable.

25 The example hereinbelow illustrates the invention.

Example

The hydrogenation of benzene to cyclohexane has an 30 exotherm of $\Delta H = -214 \text{ kJ/mol}$.

The benzene hydrogenation product equilibrates to between cyclohexane and methylcyclopentane, unless the heat of reaction is removed and a relatively low temperature is maintained.

35 Studies have also shown that the reaction is substrate-limited in that the low solubility of hydrogen in benzene and cyclohexane causes the reaction mixture to

deplete in dissolved H₂ along the catalytic layer. It is therefore advantageous to use the invention to improve the supply of dissolved hydrogen.

The benzene hydrogenation process is carried out using
5 an inventive apparatus as per Fig. 2, comprising a spiral type heat exchanger reactor as per Figures 3 and 4. To this end, the reactor passage 5 mm in width, 25 mm in depth and 960 mm in length (volume 120 ml) was packed with 8 plies of knitted catalyst fabric tape
10 prepared by first heat-treating a knitted support tape of V2A stainless steel (German material number 1.4301) at 650°C for 3 h and then vacuum-coating it with 6 nm of platinum. The amount of active component was 46 mg.
The catalyst-packed heat exchanger reactor was
15 installed in the apparatus depicted in Fig. 3. After purging with nitrogen and reduction of the catalyst with hydrogen at 80°C for 2 h, benzene was pumped via the feed line 4 into the cyclohexane-filled liquid circulation system. The reaction parameters were
20 p = 20 bar, T = 90°C and a superficial liquid and hydrogen velocity of 400 m³/m²h.

The temperature of the reaction product was measured at the reactor outlet. A maximum temperature difference of 0.2°C was observed relative to the reaction temperature
25 setting.

A selectivity of 100% was obtained with 98% conversion. The space-time yield based on the volume of the reactor passage was 0.5 kg/(l·h).

30

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We claim:-

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1. Apparatus for carrying out reactions involving a gaseous phase, a liquid phase and a solid phase, comprising

10

- a dispersing element (6) for dispersing a gas phase in a liquid phase to generate a reaction fluid,

15

- at least one reactor (1) which possesses an inlet (31, 41), an outlet (43) and a reactor space bounded by heat-removing walls which are spaced apart substantially uniformly along the main flow axis of the reaction fluid, and which is fitted with catalyst-coated metal fabric (20, 32), and

20

- a feed line (7) which routes the reaction fluid from the dispersing element (6) to the reactor inlet (31, 41) and is sufficiently short that the degree of dispersion of the reaction fluid does not substantially change in the course of the passage through the feed line.

25

2. Apparatus as claimed in claim 1, wherein the metal fabric (20, 32) is woven metal fabric.

3. Apparatus as claimed in claim 1, wherein the metal fabric (20, 32) is knitted metal fabric.

30

35 4. Apparatus as claimed in claim 1, wherein the dispersing element (6) is a liquid jet gas compressor.

RECORDED IN U.S. PATENT AND TRADEMARK OFFICE

5. Apparatu[REDACTED] as claimed in claim [REDACTED] wherein the reactor (1) is constructed as a heat exchanger.
6. Apparatus as claimed in claim 5, wherein the reactor (1) is constructed as a plate type heat exchanger.
5
7. Apparatus as claimed in claim 5, wherein the reactor (1) is constructed as a spiral type heat exchanger.
10
8. Apparatus as claimed in claim 5, wherein the walls in the reactor are spaced from 1 to 30 mm apart.
- 15 9. Apparatus as claimed in claim 5, wherein the walls in the reactor are spaced from 2 to 20 mm apart.
10. Apparatus as claimed in claim 5, wherein the walls in the reactor are spaced from 4 to 10 mm apart.
20
11. A process for carrying out reactions involving a gaseous phase, a liquid phase and a solid phase, which comprises the steps of
25
 - generating a reaction fluid by dispersing a gas phase in a liquid phase,
 - passing the generated reaction fluid through a reactor whose reactor space is equipped with woven or knitted metal fabrics coated with catalyst,
30
 - transferring the heat of reaction at the walls which bound the reactor space, and
- 35
 - separating the reaction fluid into gas phase and liquid phase.

12. A process as claimed in claim 11, operated with separate partial recycling of gas phase and/or liquid phase.

5

13. A process as claimed in claim 11, wherein the superficial liquid velocity in the reactor is from 100 to 600 m³/(m²·h).

10 14. A process as claimed in claim 11, wherein the superficial gas velocity is from 0.5 to 15 cm/s.

15 15. A process as claimed in claim 11, wherein the reaction fluid in the reactor is under a pressure of from 0.1 to 200 bar.

16. A process as claimed in claim 11, wherein the reaction fluid in the reactor has a temperature of from 25 to 250°C.

20

BASF Aktien
Gesellschafty 29, 2000
NAE19980416 US IB/UK/fey**Abstract**

The invention relates to a process and apparatus for the isothermal operation of heterogeneously catalyzed reactions involving at least three phases in the form of a gaseous phase, a liquid phase and a solid phase. The invention provides apparatus for carrying out reactions involving a gaseous phase, a liquid phase and a solid phase, comprising (i) a dispersing element for dispersing a gas phase in a liquid phase to generate a reaction fluid, (ii) at least one reactor which possesses an inlet, an outlet and a reactor space bounded by heat-removing walls which are spaced apart substantially uniformly along the main flow axis of the reaction fluid, and which is fitted with catalyst-coated metal fabric, and (iii) a feed line which routes the reaction fluid from the dispersing element to the reactor inlet and is sufficiently short that the degree of dispersion of the reaction fluid does not substantially change in the course of the passage through the feed line.

United States Patent [19]

Arganbright et al.

[11] Patent Number: **4,950,834**

[45] Date of Patent: **Aug. 21, 1990**

[54] ALKYLATION OF ORGANIC AROMATIC COMPOUNDS IN A DUAL BED SYSTEM

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[21] Appl. No.: **385,443**

[22] Filed: **Jul. 26, 1989**

[51] Int. Cl.⁵ C07C 2/66

[52] U.S. Cl. 585/446; 585/449;
585/450; 585/467

[58] Field of Search 585/449, 450, 467, 446

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Primary Examiner—Glenn Calderola

Assistant Examiner—William C. Diemler

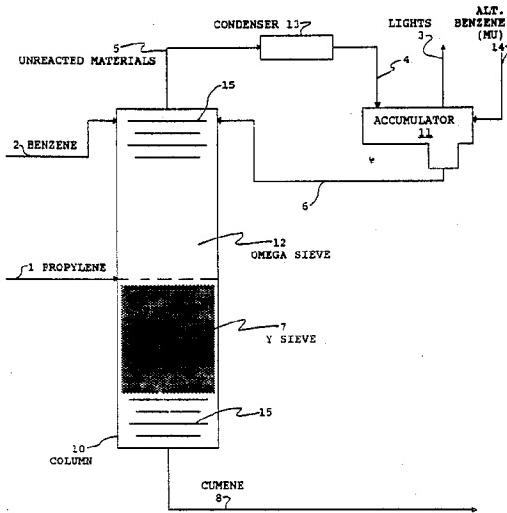
Attorney, Agent, or Firm—Kenneth H. Johnson

[57]

ABSTRACT

Cumene is produced in a catalytic distillation column reactor having an upper bed of Omega type molecular sieve catalyst and a lower bed of Y type molecular sieve catalyst. Benzene and propylene are reacted in the upper bed where the Omega type sieve is more selective to cumene than the Y type sieve. Part of the reaction mixture flows down the column to the Y bed where benzene reacts with any unreacted propylene to produce cumene. Additionally, benzene reacts with dipropylbenzene in the Y bed to produce more cumene. Cumene is recovered as bottoms product and unreacted benzene recovered as overheads where it may be returned as reflux to the column to control the mole ratio of benzene to propylene.

22 Claims, 2 Drawing Sheets



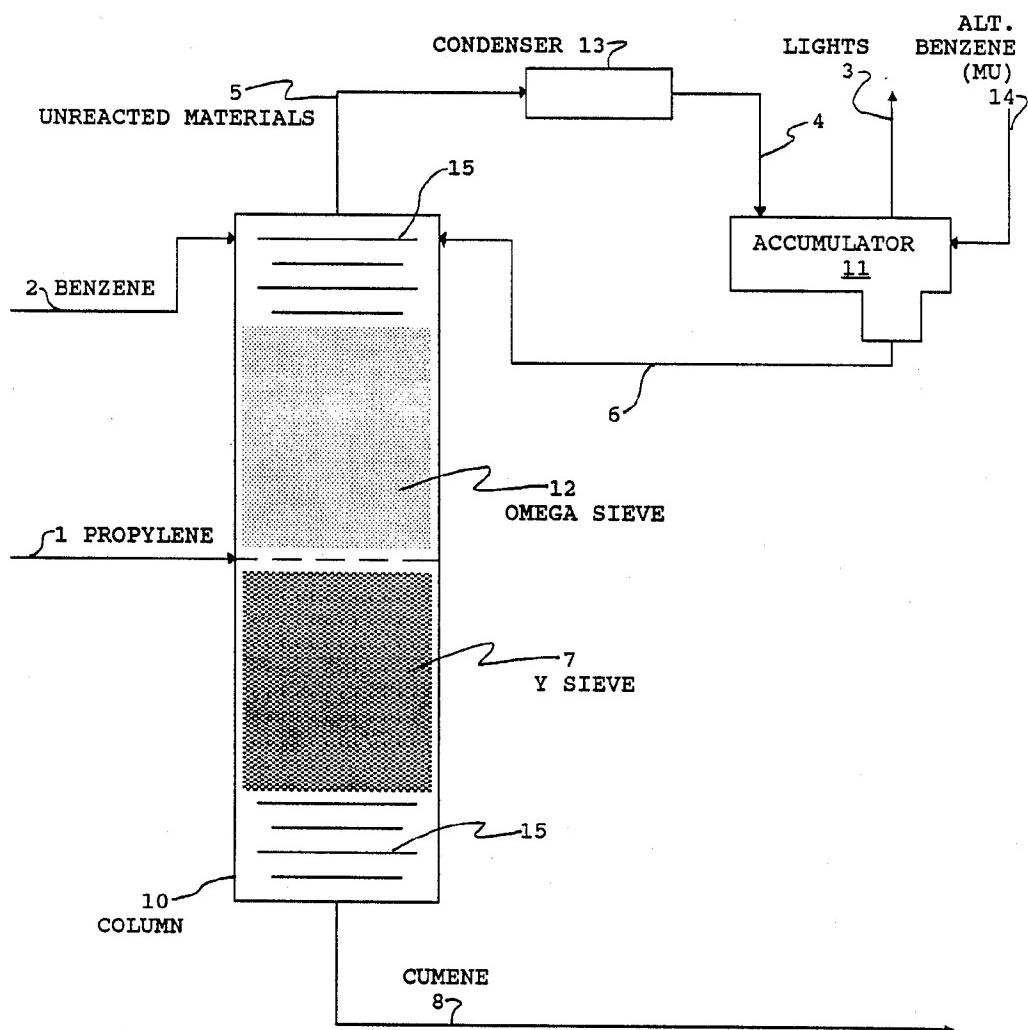


FIG. 1

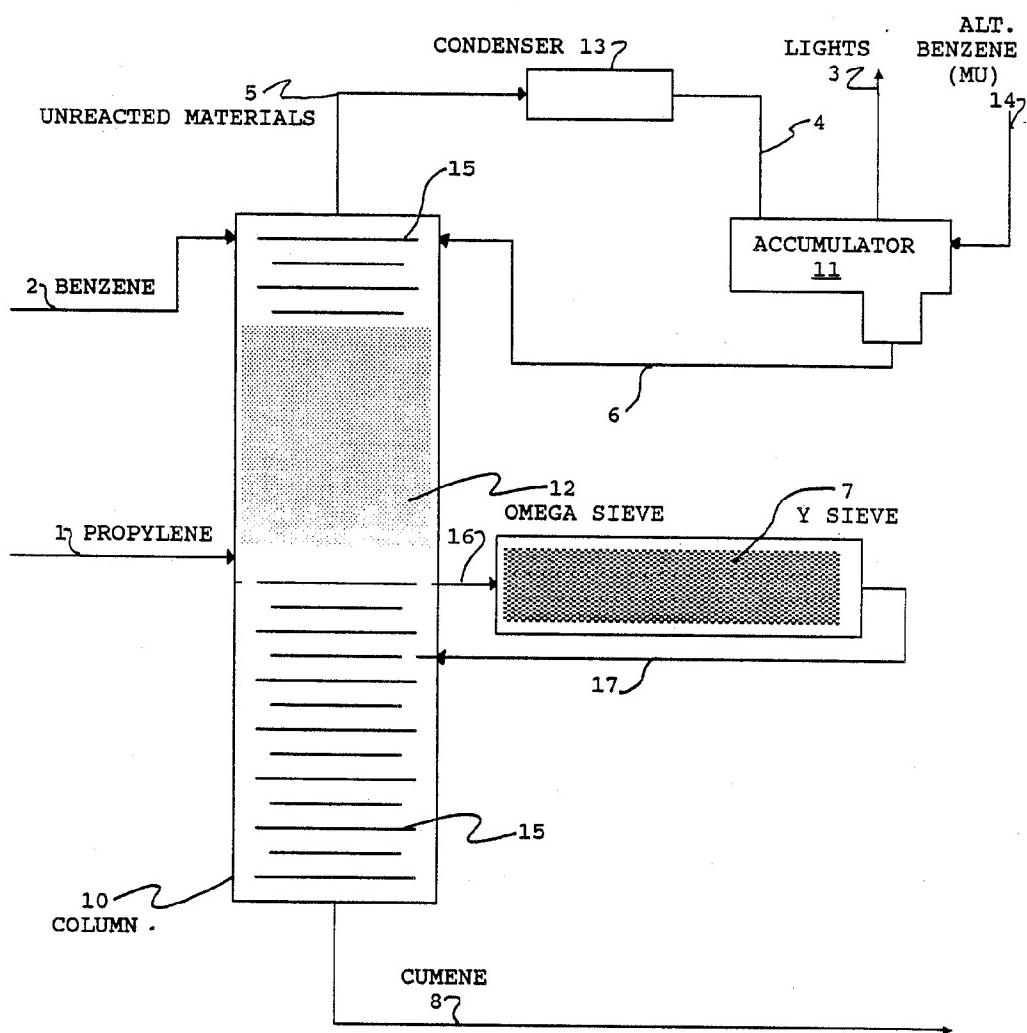


FIG. 2

ALKYLATION OF ORGANIC AROMATIC COMPOUNDS IN A DUAL BED SYSTEM

BACKGROUND OF THE INVENTION

1. Field of The Invention

The present invention relates to a process for the alkylation of organic aromatic compounds. In particular the invention relates to the alkylation of benzene with propylene to produce cumene. More particularly the invention relates to a process for the concurrent alkylation and distillation of reaction components (reactants and products) in a catalyst bed wherein the catalyst also serves as the distillation structure. Most particularly, the catalyst utilized is a dual bed of Omega and Y type molecular sieves.

2. Related Art

Recently a new method of carrying out catalytic reactions has been developed, wherein the components of the reaction system are concurrently separable by distillation, using the catalyst structures as the distillation structures. Such systems are described variously in U.S. Pat. Nos. 4,215,011; 4,232,177; 4,242,530; 4,302,356; 4,307,254; 4,336,407; 4,439,350; 4,443,559; and 4,482,775 commonly assigned herewith.

Briefly, a structure described there is a cloth belt with a plurality of pockets spaced along the belt, which is then wound in a helix about a spacing material such as stainless steel knitted mesh. These units are then disposed in the distillation column reactor. In addition, commonly assigned U.S. Pat. Nos. 4,443,559 and 4,250,052 disclose a variety of catalyst structures for this use and are incorporated herein.

Ethylbenzene and cumene are currently produced by the reaction of benzene and the respective olefin, i.e., ethylene and propylene by acid catalysis. In some known processes the catalyst is highly corrosive and has a relatively short life, e.g., AlCl_3 , H_3PO_4 on clay, BF_3 on alumina, and others require periodic regeneration, e.g., molecular sieves. The exothermicity of the reaction and the tendency to produce polysubstituted benzene require low benzene conversions per pass with large volume recycle in conventional processes. Advantages of the present invention are that the catalysts are not highly corrosive and may not require periodic regeneration, the heat of reaction is used efficiently, only low volume is required and the feed ratios can approach unity.

A catalytic distillation process for the production of cumene is disclosed in co-pending U.S. patent application Ser. No. 07/122,485, where a single bed of either acid cation exchange resin or a type Y molecular sieve catalyst was used.

The inventors herein have discovered a process for the alkylation of benzene with propylene that takes advantage of the higher selectivity of the Omega type sieves while reducing the amount of undesirable olefins in the cumene product.

SUMMARY OF THE INVENTION

Briefly, the present invention is a process for the alkylation of benzene by contacting the benzene and propylene in a distillation column reactor containing a dual bed of molecular sieve catalyst in a reaction distillation zone thereby catalytically reacting the benzene and propylene to produce an cumene product and concurrently in said fixed bed fractionating the resultant cumene product from the unreacted materials. The

molecular sieve catalyst in each bed provides both the catalytic sites and the distillation sites. The cumene, having a higher boiling point, is withdrawn (rom the distillation column reactor at a point below the fixed bed and unreacted benzene and propylene (if any) may be taken off as an overhead.

Specifically the two catalyst are arranged so that the initial reaction of propylene and benzene occurs in the Omega molecular sieve bed and a reaction mixture, corresponding to the mid reflux in a single bed catalytic reaction distillation system for the reaction, is directed to a type Y molecular sieve where the reaction is completed. In other words reflux that would normally go back to the Omega bed instead goes to the type Y bed.

The feed to the type Y bed may be characterized as containing benzene, cumene, isopropylene, dipropylbenzene, tripropylbenzene, propylene dimers and other oligomers. Depending on the nature of the feed, there may be other materials present which may have some bearing on the long term or overall operation of the process, but are not of direct concern to the present invention. In addition to the apparent reaction of the dimers and/or other oligomers there is a degree of transalkylation occurring that favors the cumene produc-

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic representation of a preferred embodiment of one species of the present invention for producing cumene with the catalysts in the reactor distillation column.

FIG. 2 is a schematic representation of an alternative embodiment of one species of the present invention for producing cumene wherein the type Y catalyst is not in the reactor distillation column.

DETAILED DESCRIPTION OF THE INVENTION

The Omega type molecular sieve catalyst has been found to be more selective for the production of cumene than the type Y. However, the bottoms cumene product contains an unsatisfactory amount of heavier boiling olefins. This is believed to be due to the small pore size of the Omega catalyst as compared to the type Y sieve. The smaller pore size may not allow the dimerized propylene to react with the benzene, leaving it available to react with other propylene to produce the unwanted higher boiling olefins found in the cumene product.

The dual bed system provides the additional advantage of carry over of benzene into the lower bed of Y sieve along with cumene and dimerized propylene. In that portion of the reactor the dimerized propylene can react with the benzene present and reduces the amount of unwanted heavier boiling olefins in the cumene product. Thus the higher yield of the Omega catalyst is obtained with reduced oligomerization.

The molecular sieve catalyst packing is of such a nature as to allow vapor flow through the bed, yet provide a sufficient surface area for catalytic contact as described in the previously noted U.S. Pat. Nos. 4,215,011 and 4,302,356 which are incorporated herein in their entirety. The two distinct beds are comprised of an Omega type molecular sieve and a Y type molecular sieve. The Omega sieve is disposed in the upper one-third to one-half of the distillation column reactor with the Y sieve occupying the lower portion of the reactor.

The propylene feed is added either below the lower bed of catalyst or between the two beds, but preferably between the two beds. The benzene is preferably added above the Omega sieve and may be conveniently added to the reflux as make-up or fed separately. In order to achieve high selectivity toward monosubstitution (which is a preferred aspect of the present invention), there is a large excess of benzene to propylene in the reactor in the range of 2 to 100 moles of benzene per mole of propylene. The net molar feed ratio of benzene to propylene may be close to 1:1, but the system is operated so as to maintain a substantial molar excess of benzene to propylene in the reaction zone. The cumene product is the highest boiling material and is separated in the lower portion of the column usually as bottoms. The benzene is the second highest boiling component (excluding inerts) as noted above, however, by operating with a large excess of benzene, the major portion of the propylene is reacted, thereby reducing the separation and recovery problems.

The success of catalytic distillation lies in an understanding of the principles associated with distillation. First, because the reaction is occurring concurrently with distillation, the initial reaction product is removed from the reaction zone as quickly as it is formed. The removal of the alkylation product minimizes polysubstitution and decomposition of the alkylation product. Second, because the benzene is boiling, the temperature of the reaction is controlled by the boiling point of that component at the system pressure. The heat of the reaction simply creates more boil up, but no increase in temperature. Third, the reaction has an increased driving force because the reaction products have been removed and cannot contribute to a reverse reaction (Le Chatelier's Principle).

As a result, a great deal of control over the rate of reaction and distribution of products can be achieved by regulating the system pressure. Also, adjusting the through-put (residence time a liquid hourly space velocity) gives further control of product distribution and degree of olefin conversion.

The temperature in the reactor is determined by the boiling point of the liquid mixture present at any given pressure. The temperature in the lower portions of the column will reflect the constitution of the material in that part of the column, which will be higher than the overhead; that is, at constant pressure a change in the temperature of the system indicates a change in the composition in the column. To change the temperature the pressure is changed. Temperature control in the reaction zone is thus controlled by the pressure; by increasing the pressure, the temperature in the system is increased, and vice versa. It can also be appreciated that in catalytic distillation as in any distillation there is both a liquid phase (internal reflux) and a vapor phase. Thus, the reactants are partially in liquid phase which allows for a more dense concentration of molecules for reaction, whereas, the concurrent fractionation separates product and unreacted materials, providing the benefits of a liquid phase system (and a vapor phase system) while avoiding the detriment of having all of the components of the reaction system continually in contact with the catalyst which would limit the conversion to the equilibrium of the reaction system components.

Molecular sieves are porous crystalline, three-dimensional alumina-silicates of the zeolite mineral group. The crystal skeleton is composed of silicon and aluminum atoms each surrounded by four oxygen atoms to

form the crystalline structure. The term molecular sieve can be applied to both naturally occurring zeolites and synthetic zeolites. Naturally occurring zeolites have irregular pore size and are not generally considered as equivalent to synthetic zeolites. In the present invention, however, naturally occurring zeolites are acceptable so long as they are substantially pure. The balance of the present discussion shall be directed to the synthetic zeolites with the understanding that natural zeolites are considered equivalent thereto as indicated above, i.e., in so far as the natural zeolites are the functional equivalents to the synthetic zeolites.

Usually synthetic zeolites are prepared in the sodium form, that is, with a sodium cation in close proximity to each aluminum tetrahedron and balancing its charge. To date seven principal types of molecular sieves have been reported, A, X, Y, L, erionite, Omega and mordenite. The A type have relative small pore size. By the term ore size is meant the effective pore size (diameter) rather than the free pore size (diameter). Types X and Y have larger pore size (approximately 10 Å.) and differ as to the range of ratio of Al_2O_3 to SiO_2 as:

Type X— Al_2O_3 /2.0–3.0 SiO_2

Type Y— Al_2O_3 /3.0–6.0 SiO_2

Type L and the other types listed have still higher ratios of SiO_2 to Al_2O_3 . Of particular interest is the Omega type which has an Al_2O_3 to SiO_2 ratio of 1:5 to 1:12.

The mole sieve catalysts employed in the present invention are the acid form mole sieves or exhibit acidic characteristics. The acid form of the mole sieves is commercially available, but also may be prepared by treating the mole sieves with acid to exchange Na for hydrogen. Another method to produce the acid form is to treat the mole sieve with decomposable cations (generally ammonium ions) to replace Na with the decomposable ions and thereafter to heat the mole sieve to decompose the cation leaving the acid form. Generally the Na form mole sieve is treated with ammonium hydroxide to remove the Na and thereafter the mole sieve is heated to a temperature of about 350° C. to remove the ammonia. The removal of Na^+ ions with NH_4^+ is more easily carried out than with multivalent ions as described below and these catalysts are generally more active, but less stable to heat than the multivalent cation exchange forms. Mole sieves, which have had their alkali metal reduced to low levels by partial treatment with NH_4^+ and partial multivalent metal cation exchange, possess increased activity and increased stability. In addition to mole sieves which are acidic according to the Bronsted Theory those mole sieves which exhibit acidic characteristics under the Lewis Theory, for example, calcium exchanged mole sieves, are suitable for the present reaction. By exchanging the univalent cations (e.g.) Na^+ with multivalent cations, strong ionic activity is imparted. The ratio of SiO_2 : Al_2O_3 Valence and radius of the cation and the extent of exchange all affect the catalyst activity. In general activity increases with (1) increased SiO_2 Al_2O_3 ratio, (2) decreased cation radius and an increase in cation valence. The effect of replacing univalent ions (e.g. Na^+) with bivalent (e.g. Ca^{++}) is much greater than replacing the bivalent ions with cations of greater valence.

The various types of mole sieves having reduced alkali metal content are characterized as the acid form molecular sieve and are all contemplated as useful in the present invention.

It would appear that the pore size within the crystal lattice may affect the selectivity. According to one theory of molecular sieve catalytic activity, zeolite catalysis occurs primarily inside the uniform crystal cavities, consequently zeolitic catalyst activity depends on the number of aluminum atoms in the crystal and thus on the chemical composition of the crystal. Moreover, these catalytic sites are fixed within the rigid structure of the crystal, so that access to site can be altered by altering the structure of the crystal.

The acid form mole sieves are generally produced and available as particles in the range of <10 micron powders) to 0.2 inch in diameter (beads).

In this form the mole sieves form too compact a bed and will not function adequately in a distillation, since there is a very large pressure drop through the bed and the free flow of internal reflux and rising vapor is impeded. Mole sieves in the shape of conventional distillation structures, such as rings, saddles, and the like may be used in the present invention. The particulate mole sieves may be employed by enclosing them in a porous container such as cloth, screen wire or polymeric mesh. The material used to make the container must be inert to the reactants and conditions in the reaction system. The cloth may be any material which meets this requirement such as cotton, fiber glass, polyester, nylon and the like. The screen wire may be aluminum, steel, stainless steel and the like. The polymer mesh may be nylon, teflon or the like. The mesh or threads per inch of the material used to make the container is such that the catalyst is retained therein and will not pass through the openings in thermal aerial. Particles of about 0.15 mm size or powders may be used and particles up to about $\frac{1}{4}$ inch diameter may be employed in the containers.

The container employed to hold the catalyst particles may have any configuration, such as the pockets disclosed in the commonly assigned patents above or the container may be a single cylinder, sphere, doughnut, cube, tube or the like.

Each container containing a solid catalytic material comprises a catalyst component. Each catalyst component is intimately associated with a spacing component which is comprised of at least 70 volume % open space up to about 95 volume % open space. This component may be rigid or resilient or a combination thereof. The combination of catalyst component and spacing component form the catalytic distillation structure. The total volume of open space for the catalytic distillation structure should be at least 10 volume % and preferably at least 20 volume % up to about 65 volume %. Thus desirably the spacing component or material should comprise about 30 volume % of the catalytic distillation structure, preferably about 30 volume % to 70 volume %.

Resilient materials are preferred. One suitable such material is open mesh knitted stainless wire, known generally as demister wire or an expanded aluminum. Other resilient components may be similar open mesh knitted polymeric filaments of nylon, teflon and the like. Other materials such as highly open structures foamed material, e.g., reticulated polyurethane foam (rigid or resilient) may be formed in place or applied around the catalyst component. In the case of larger catalyst components such as from about $\frac{1}{4}$ inch to $\frac{1}{2}$ pellets, spheres, pills and the like each such larger component may be individually intimately associated with or surrounded by the spacing component as described above. It is not essential that the spacing component entirely cover the

catalyst component. It is only necessary that the spacing component intimately associated with the catalyst component will act to space the various catalyst components away from one another as described above. Thus, the spacing component provides in effect a matrix of substantially open space in which the catalyst components are randomly but substantially evenly distributed.

A preferred catalytic distillation structure for use herein comprises placing the mole sieve particles into a plurality of pockets in a cloth belt, which is supported in the distillation column reactor by open mesh knitted stainless steel wire by twisting the two together in a helical form. This allows the requisite flows and prevents loss of catalysts. The cloth may be any material which is inert in the reaction. Cotton or linen are useful, but fiber glass cloth or "Teflon" cloth are preferred.

In the following examples the catalyst packing consisted of bags in the form of a fiber glass cloth belt approximately six inches wide with narrow pockets approximately $\frac{1}{4}$ inch wide sewn across the belt. The pockets are spaced about $\frac{1}{4}$ inch apart. These pockets are filled with the catalyst particles to form approximately cylindrical containers, and the open ends are then sewn closed to confine the particles. This belt is then twisted into a helical form to fit inside the column. Twisted in with the belt is also a strip of an open mesh knitted stainless steel wire, which serves to separate the mole sieve filled cloth pockets and provide a passage for vapor flow.

The wire mesh provides the support for the catalyst (belt) and provides some degree of vapor passage through the catalyst particles which otherwise form a very compact bed which has a high pressure drop. Thus, the down flowing liquid is in intimate contact with the rising vapors in the column.

In commercial-scale operations, it is contemplated, catalyst packing would be made up of alternating layers of mole sieve filled cloth belts similar to the ones described above, and a spacing material which could be of any convenient, suitable substance, such as a corrugated wire screen or wire cloth or a knitted wire mesh. The layers would be arranged vertically or horizontally. For simplicity of fabrication and for better distribution of vapor flow passages, a vertical orientation is preferred. The height of a section of this packing should be of any convenient dimension, from a few inches to several feet. For ease of assembly and installation, the packing would be made into sections of the desired shape and size, each section fastened together with circumferential bands of tie wires depending on its size and shape. A complete assembly in a column would consist of several sections, arranged in layers, with possibly the orientation of the catalyst-filled belts turned at right angles in successive layers to improve liquid and vapor flow distribution.

The preferred arrangement of the two different type mole sieve beds is to have a lower bed of type Y filling the lower one-half to two-thirds of the column with the Omega sieve filling the remainder. Although in two distinct beds, the method described above would be applied to each bed.

FIG. 1 illustrates one embodiment of the present invention. Referring to the drawing, distillation column reactor 10 is divided into two sections. In the lower section 7 the catalyst packing (catalytic distillation structures) are positioned as described. Linde molecular sieve LZ-Y82 1/16" (Union Carbide Corp.) is deposited in the pockets of fiber glass belts and formed in to a

helix with stainless steel mesh as described. In the upper section 12 Union Carbide Omega type sieve is likewise loaded into the reactor 10. Conventional distillation trays or structures 15 are positioned above and below the catalyst beds.

The lower portion as well as the upper of the column may contain conventional distillation column structure (trays or inert packing) to achieve the desired final separation in the lower and upper sections of the column. In the drawing the benzene is indicated to be introduced into the column 10 via flow line 2 into the upper bed 12 it may be conveniently added as makeup into reflux accumulator 11. The propylene is fed to the column via flow line 1 at about the mid point between the two catalyst beds 7 and 12 or below the lower catalyst bed (not shown) for better mixing. The propylene may also be fed at several points to reduce the concentration at any one location in the catalyst zone, thus reducing oligomerization as a side reaction. The reaction is exothermic and initiated by contacting the two reactants in the catalyst packing. Cumene is the principal reaction product in the Omega bed 12, however dimerized propylene is also produced along with some dipropylbenzene. Since complete separation of the reaction products and benzene does not occur in the Omega bed 12, cumene, small amounts of propylene, dimerized propylene, dipropylbenzene (tripropylbenzene is also present) and benzene flow down the column into the Y bed where any propylene and the dimerized propylene may react with the benzene to produce additional cumene and prevent formation of undesired higher boiling olefins. Additionally, the dipropylbenzene is transalkylated with unreacted benzene to produce additional cumene.

The cumene product is higher boiling than benzene and propylene and is recovered via flow line 8 as a bottoms product. The feed of propylene is adjusted such that there is a molar excess of benzene in the reactor, such that the overhead 5 is primarily benzene, the propylene having been almost totally reacted. In addition to benzene and some propylene other lights go off overhead. The overhead is passed to condenser 13 which is operated to condense substantially all of the benzene which passes via flow line 4 to accumulator 11 and hence, by reflux via flow line 6 to column 10. The benzene used in the reaction and lost with the lights (which exit accumulator 11 via 3) is made up by fresh benzene feed through flow line 2 or alternatively to accumulator 4 through flow line 14.

FIG. 2 shows the embodiment wherein the Omega catalyst is in the distillation reactor as described before, except that the type Y sieve is outside of the column. A portion (any amount up to 100%) of the mid-reflux of the column is removed via line 16 and passed through the type Y sieve then back via 17 to the lower portion of the column containing conventional distillation trays 15.

Such conventional items as valves, reboilers, slip streams, etc. are not shown, but would be obvious expedients to those setting up such equipment.

The mole ratio of benzene to propylene in the column may be in the range of 2 to 100:1, preferably 2 to 50:1 and more desirably about 2 to 10:1. The greater the excess of benzene the more the selectivity to the mono-substituted product is improved. Alkylation is forced to completion, since the simultaneous and concurrent fractionation and removal of the alkylation product from the distillation column reactor does not allow the products to contribute to the reverse reaction (Le Chatelier's

Principle). However, very large molar excesses of benzene require a very high reflux ratio, and a low unit productivity. In this reaction the propylene is the most volatile component and it is desirable to react it rather than have some carried off overhead. The presence of propylene or other lower boiling olefin in the tower with benzene will result in a small but detectable temperature depression in the tower where such lower boiling olefins are present as entities and unreacted. As the propylene is reacted with benzene, the depressing effect is diminished and furthermore, the reaction, which is exothermic, also diminishes the effect. The magnitude of the temperature depression immediately above the propylene feed is a measure of the concentration of propylene in the system, that is, the larger the concentration of the propylene, the greater the depression of the temperature where the benzene and propylene are initially together and yet unreacted. For this particular system the concentration of propylene to provide a given temperature depression can be determined and plotted. Thus, by maintaining a specific temperature at the point of maximum temperature depression by adjusting the propylene feed, a given ratio of propylene to benzene can be maintained in a simple and expedient manner. More significantly, the maintenance of the depression at a given temperature can assure that substantially all of the propylene will be reacted prior to the end of the catalyst bed and overhead exit, if the corresponding propylene concentration has been determined to produce that effect.

The present alkylation reaction can be carried out at sub-through super atmospheric pressure, e.g., 0.20 to 40 atmospheres. The temperature will vary depending on the reactants and product. Furthermore, the temperature along the column will be as in any distillation column, the highest temperature will be in the bottom and the temperature along the column will be the boiling point of the composition at that point in the column under the particular conditions of pressure. Moreover, the exothermic heat of reaction does not change the temperature in the column, but merely causes more boil up. However, the temperatures within the column with the above considerations in mind will generally be in the range of 50° C., e.g. 70° C. to 500° C. and more preferably in the range of about 80° C. to 300° C. at pressures of 0.5 to 20 atmospheres.

In a preferred embodiment the alkylation reaction is carried out by increasing the liquid level in the reaction distillation zone containing the Omega type molecular sieve. This is achieved by a liquid flow restrictor between the reaction distillation zone and the lower distillation zone. That is, the vapor from below may rise up to (and through) the reaction distillation zone as in a conventional or prior operation but a portion of the liquid is maintained there. If a single distillation column reactor is used, a conventional distillation tray with the downcomer area blocked is located between the reaction distillation zone and the distillation zone. A by pass line for liquid flow is provided about the tray and a valve is provided in the liquid flow conduit to restrict liquid downflow and thereby to build up a liquid level above that tray just below the catalyst bed. Alternatively a perforated plate may be used to support the catalyst and cause a liquid pressure drop in the column thus building up a level in the catalyst. If the two column system is used, then a valve or other restriction means is placed in the liquid flow line between the two columns.

While the particular position of the liquid level has been described above to be at the lower end of the reaction distillation zone, it could just as easily be placed anywhere in the catalyst bed depending upon the desired reactions.

The term "liquid level" is used herein to mean an increased density of the material in the reaction distillation zone over that of a pure distillation as distinguished to a continuous liquid phase. The phase system as present in the reaction distillation zone is physically a froth. This is the result of the vapor traveling up through the liquid retained in the zone.

Another way of viewing this is that in normal distillation there is a vapor with liquid (internal reflux) trickling down through the vapor and contacting the catalyst whereas in the present "flooded" system the vapor is traveling up through a liquid phase to create the froth or foam.

Hence in essence the benefits of the distillation are still obtained, i.e., separating the various components by the distillation whereas the increased liquid volume in contact with the catalyst improves the synthesis reaction. This method of operation is more fully described in commonly owned U.S. patent application Ser. No. 07/328,487, which is incorporated herein by reference.

EXAMPLE 1

The reactor was a 3 inch diameter pilot distillation tower having a 29 foot section packed with molecular sieve catalyst contained in glass cloth pockets twisted with demister wire as described above.

Two runs were carried out under substantially the same conditions using a Y type and Omega molecular sieve respectively. The results of the two runs are set out in Table I. Although the Omega catalyst showed better selectivity for cumene, the high content of olefin (indicated by the high bromine number) made the product commercially unacceptable.

TABLE I

Run	1	2
Catalyst	Y-82*	Omega
Mid-reflux Analysis, Wt. %		
Benzene	78.09	78.00
Cumene	17.01	18.12
DIPB	3.78	2.86
TIPB	0.29	0.15
Wt. ratio,		
Cumene/DIPB + TIPB	4.2	6.0
Selectivity		
Benzene to cumene	85.1	89.1
Bromine No. of cumene product	6	>40

*Product of Union Carbide Corporation

EXAMPLE 2

Samples of the mid internal reflux from runs using the Omega sieve were collected and combined and fed over a Y-82 mole sieve catalyst (Linde) at 310° F. (LHSV 3 to 11) in a $\frac{1}{2}$ inch isothermal reactor (as represented by FIG. 2). Analysis by chromatography and bromine number indicated that the olefins were removed to an acceptable level by the use of the Y-82 catalyst. Bromine titration is an accepted indication of the amount of olefins in a liquid. The propylene remaining in the mid-reflux sample was reacted to produce more cumene, and high cumene/dipropylbenzene ratio was improved due to some transalkylation obtained with the Y-82 as shown in Table II below.

TABLE II

	Feed	Product 3 LHSV	Product 11 LHSV
5	Cumene, wt. % ¹	90	95
	Cumene/dipb wt ratio	9	20
Cumene X 100 Cumene + DIPB + TIPB + HE (excludes benzene)			

10 Complete analysis of the feed and product are given in Table III below. The bromine number of the product was 4 which compares with the bromine number of 102 obtained on tower bottoms when using the Omega sieve alone.

TABLE III

REACTION OF OMEGA CATALYZED MID-REFLUX OVER Y 82 CATALYST			
Compound	Feed, wt. %	Product 3 LHSV	Product 11 LHSV
$C_3=$	0.258	0.000	0.000
Unknown	0.006	0.000	0.014
Benzene	84.488	82.784	83.013
Unknown	0.158	0.132	0.155
Toluene	0.018	0.030	0.017
Unknown	0.124	0.024	0.012
Unknown	0.032	0.031	0.024
Unknown	0.003	0.024	0.000
Cumene	13.386	16.152	15.437
DIPB	0.460	0.543	0.763
DIPB	0.192	0.000	0.016
DIPB	0.766	0.280	0.433
TIPB	0.052	0.000	0.016
Bromine No.	>100	4	12.6

The invention claimed is:

1. A process for the production of cumene by the alkylation of benzene with propylene comprising the steps of:
 - (a) feeding a stream containing propylene into a distillation reactor column at a point below a bed of Omega molecular sieve catalyst prepared as distillation structures;
 - (b) feeding benzene into said distillation reactor column at a point above said bed of Omega molecular sieve catalyst;
 - (c) concurrently in said distillation reactor column:
 - (1) reacting a portion of said propylene with said benzene within said bed of Omega molecular sieve catalyst to form a reaction mixture containing cumene, unreacted benzene, unreacted propylene and other reaction products, said other reaction products including dipropylbenzene and dimerized propylene, and
 - (2) fractionally distilling said reaction mixture within said bed of Omega molecular sieve catalyst to partially separate said cumene from said unreacted benzene, said unreacted propylene and said other reaction products forming a liquid phase containing cumene, unreacted benzene, unreacted propylene and other reaction products, and a gaseous phase substantially free of said cumene;
 - (d) contacting said liquid phase with a bed of zeolite Y molecular sieve catalyst to preferentially react said unreacted benzene contained therein with said unreacted propylene and propylene dimer and said dipropylbenzene contained therein to form additional cumene;

(e) after contact with said zeolite Y molecular sieve catalysts, fractionally distilling said liquid phase in said distillation reactor column into said bed of Omega molecular sieve catalyst;

(f) withdrawing cumene from a point below said bed of Omega molecular sieve catalyst; and

(g) withdrawing unreacted benzene and unreacted propylene at a point above said bed of Omega molecular sieve catalyst.

2. The process according to claim 1 wherein said zeolite Y molecular sieve catalyst is positioned in said distillation reactor column below said Omega molecular sieve catalyst and adapted to serve a distillation structure.

3. The process according to claim 1 wherein said zeolite Y molecular sieve catalyst is positioned outside of said distillation reactor column.

4. A process for the alkylation of benzene with propylene to produce cumene in a distillation reactor column having an upper bed of Omega molecular sieve catalyst and a lower bed of Y zeolite molecular sieve catalyst, comprising the steps of:

(a) feeding a stream containing propylene into said distillation reactor column at a point below said bed of Omega molecular sieve catalyst;

(b) concurrently feeding benzene into said distillation reactor column at a point above said bed of Omega molecular sieve catalyst;

(c) concurrently in said distillation reactor column:

(1) reacting a portion of said propylene with said benzene within said bed of Omega molecular sieve catalyst to form a reaction mixture containing cumene, unreacted benzene, unreacted propylene and other reaction products, said other reaction products including dipropylbenzene and dimerized propylene,

(2) fractionally distilling said reaction mixture within said bed of Omega molecular sieve catalyst to partially separate said cumene from said unreacted benzene, said unreacted propylene and said other reaction products forming a liquid phase containing cumene, unreacted benzene, unreacted propylene and other reaction products, and a gaseous phase substantially free of said cumene,

(3) contacting said liquid phase with said lower bed of zeolite Y molecular sieve catalyst to preferentially react said unreacted benzene contained therein with said unreacted propylene and propylene dimer and said dipropylbenzene contained therein to form additional cumene, and

(4) fractionally distilling said liquid phase in said lower bed of zeolite Y molecular sieve catalyst to separate any unreacted benzene contained therein as a vapor back up said distillation reactor column into said upper bed of Omega molecular sieve catalyst;

(d) withdrawing cumene from a point below said lower bed of zeolite Y molecular sieve catalyst; and

(e) withdrawing unreacted benzene and unreacted propylene at a point above said upper bed of Omega type molecular sieve catalyst.

5. The process of claim 4 wherein there is a molar excess of said benzene to said propylene in said distillation column reactor.

6. The process of claim 5 wherein substantially all of said propylene is reacted with said benzene.

7. The process of claim 4 wherein a portion said unreacted benzene is condensed and returned to said distillation column reactor at a point above said upper bed of Omega molecular sieve catalyst as reflux.

8. The process of claim 7 wherein make up benzene is added to said distillation column reactor in a molar ratio of benzene to propylene of 1:1 and a molar excess of benzene to propylene is maintained by said reflux.

9. The process of claim 8 wherein said make up benzene is added to said reflux.

10. The process of claim 4 wherein the temperature in said upper bed of Omega molecular sieve catalyst at the point of said propylene feed is the boiling point of said benzene at the operating pressure of said distillation column reactor.

11. The process of claim 5 wherein 2 to 100 moles of benzene per mole of propylene are present.

12. The process of claim 11 wherein from 2 to 50 moles of benzene per mole of propylene are present.

13. The process of claim 12 wherein from 2 to 10 moles of benzene per mole of propylene are present.

14. The process of claim 4 wherein the operating pressure in said distillation column reactor is in the range of 0.5 to 20 atmospheres.

15. The process of claim 14 wherein the temperature is in the range of 80° to 300° C.

16. The process according to claim 1 wherein the downward flow of internal reflux is restricted at selected points in said reaction distillation zone to maintain a liquid level above the restriction for additional contact and reaction of the liquid and distillation vapors with the Omega molecular sieve catalyst.

17. The process according to claim 2 wherein the downward flow of internal reflux is restricted at selected points in said reaction distillation zone to maintain a liquid level above the restriction for additional contact and reaction of the liquid and distillation vapors with the Omega molecular sieve catalyst.

18. The process according to claim 3 wherein the downward flow of internal reflux is restricted at selected points in said reaction distillation zone to maintain a liquid level above the restriction for additional contact and reaction of the liquid and distillation vapors with the Omega molecular sieve catalyst.

19. The process according to claim 4 wherein the downward flow of internal reflux is restricted at selected points in said reaction distillation zone to maintain a liquid level above the restriction for additional contact and reaction of the liquid and distillation vapors with the Omega molecular sieve catalyst.

20. The process according to claim 7 wherein the downward flow of internal reflux is restricted at selected points in said reaction distillation zone to maintain a liquid level above the restriction for additional contact and reaction of the liquid and distillation vapors with the Omega molecular sieve catalyst.

21. The process according to claim 8 wherein the downward flow of internal reflux is restricted at selected points in said reaction distillation zone to maintain a liquid level above the restriction for additional contact and reaction of the liquid and distillation vapors with the Omega molecular sieve catalyst.

22. The process according to claim 11 wherein the downward flow of internal reflux is restricted at selected points in said reaction distillation zone to maintain a liquid level above the restriction for additional contact and reaction of the liquid and distillation vapors with the Omega molecular sieve catalyst.

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